Ultrafast coherent diffraction imaging using X-ray free electron lasers

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Major collaborators include:

This work was performed under the auspices of the U. S. DOE by LLNL under Contract No. W-7405-ENG-4.

Acknowledgements

LLNL: Henry Chapman, Sasa Bajt, Anton Barty, Daniel Barsky, Henry Benner, Brian Bennion, Micheal Bogan, Sung-Wook Chung, Matthias Frank, Stefan Hau-Riege, Richard London, Stefano Marchesini, Tom McCarville, Alex Noy, Urs Rohner, Brent Segelke, Eberhard Spiller, Abraham Szöke, Hanna Szöke, Bruce Woods, Jennifer Alameda

Uppsala: Janos Hajdu, Gösta Huldt, Carl Caleman, Magnus Bergh, Sara Lejon, Alexandra Patriksson, David van der Spoel, Florian Burmeister, Marvin Seibert, Filipe Maia, Nicursor Timneanu

SLAC: Keith Hodgson, Sebastien Boutet

DESY: Elke Plönjes-Palm, Marion Kuhlmann, Rolf Treusch, Stefan Düsterer, Thomas Tschentscher, Jochen Schneider

TU Berlin: Thomas Möller, Christoph Bostedt, Matthias Hoener

U. Essen: Klaus Sokolowski-Tinten

Oxford: Andrea Cavalleri

UC Berkeley: Roger Falcone, Tom Allison

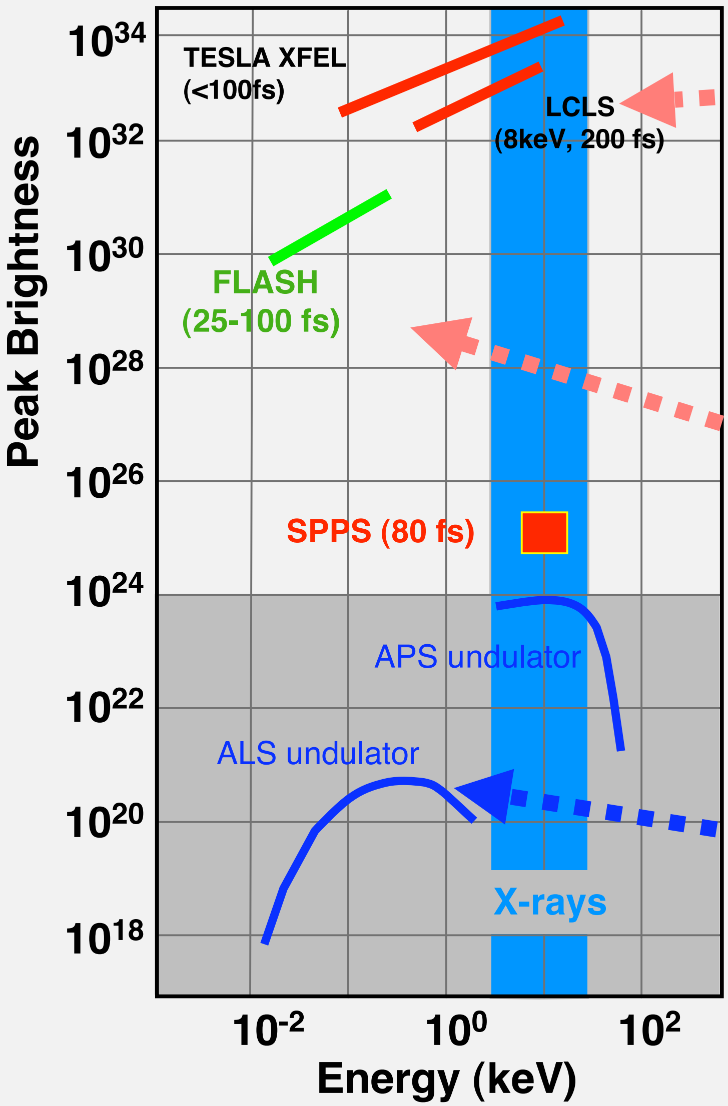
LBNL: Malcolm Howells, Congwu Cui

Stony Brook: David Shapiro, Tobias Beetz, EnjuLima, Xiaojing Huang, HuijieMiao, Andrew Stewart, Chris Jacobsen, Janos Kirz

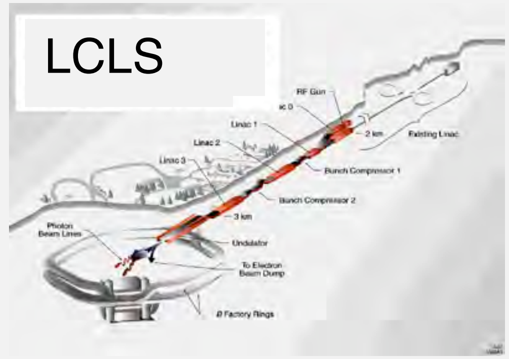
ASU: John Spence, Uwe Weierstall, Haifeng He

UC Davis: David Shapiro, Eugene Ingerman

X-ray free electron lasers are ushering in a new era in x-ray science



APS=Advanced Photon Source (ANL)
ALS=Advanced Light Source (LBNL)



operational 2008

8 keV, 200 fs, 10^{12} photons

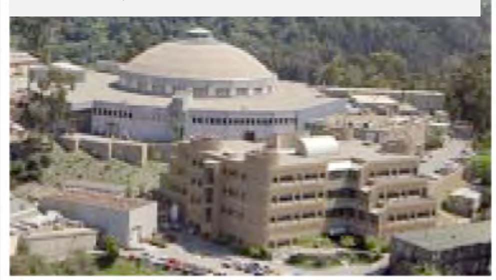
Linac Coherent Light Source, SLAC, Stanford

operational now

500 eV, 25 fs, 10^{13} photons

FLASH
DESY, Hamburg

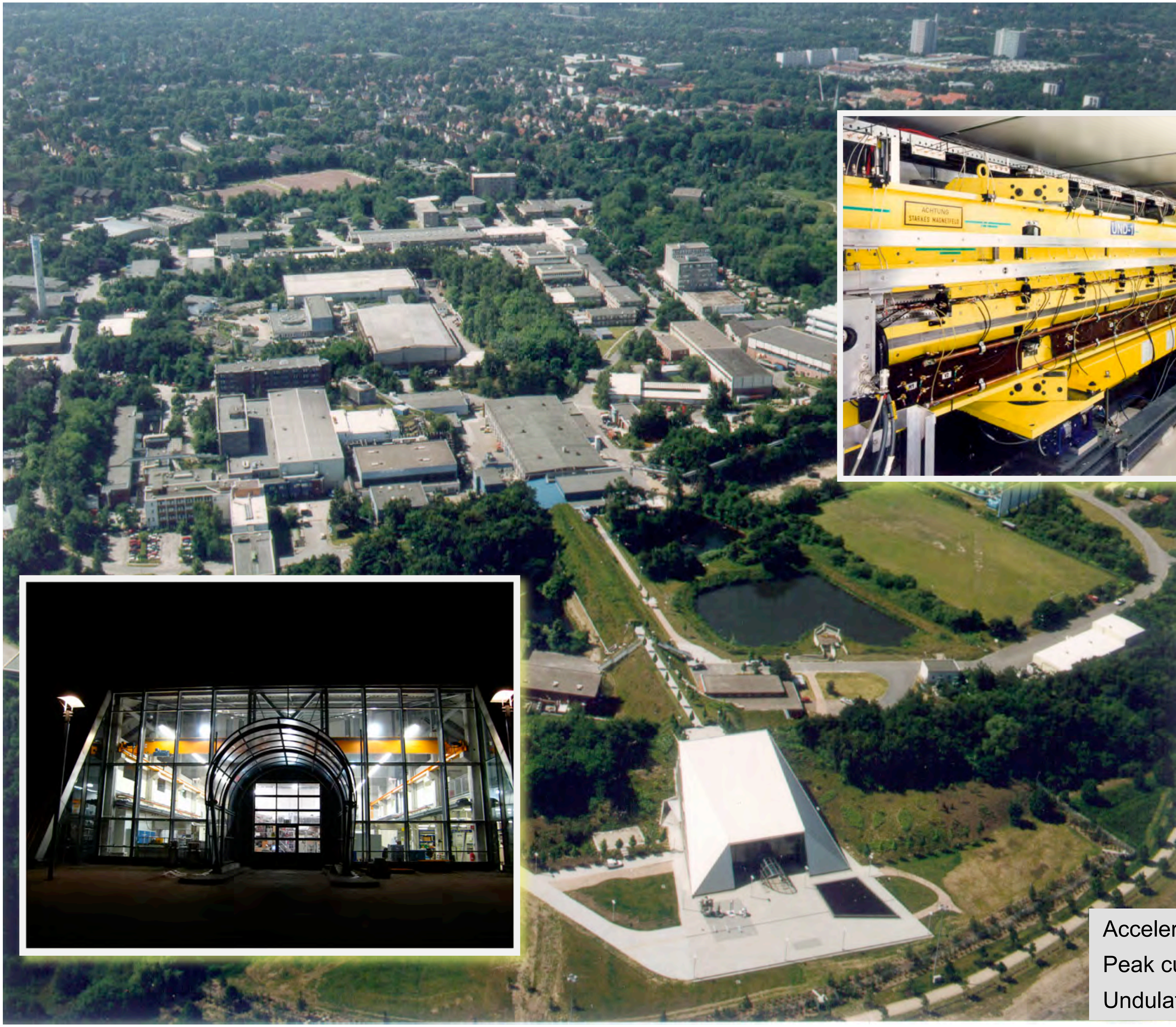
ALS, beamline 9.0.1



operational now

0.8 keV, 70 ps,
10^{10} photons/s/μm^2

Advanced Light Source undulator

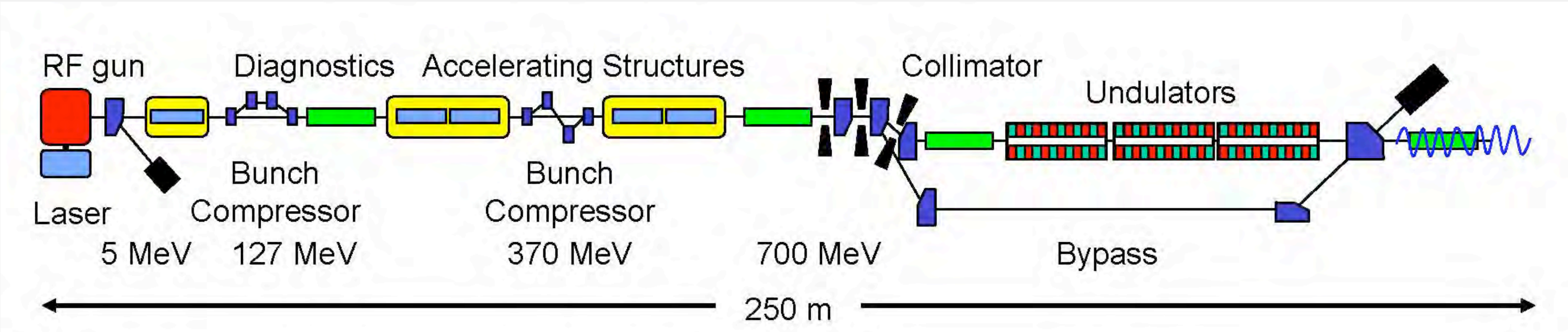


Accelerator 450 MeV

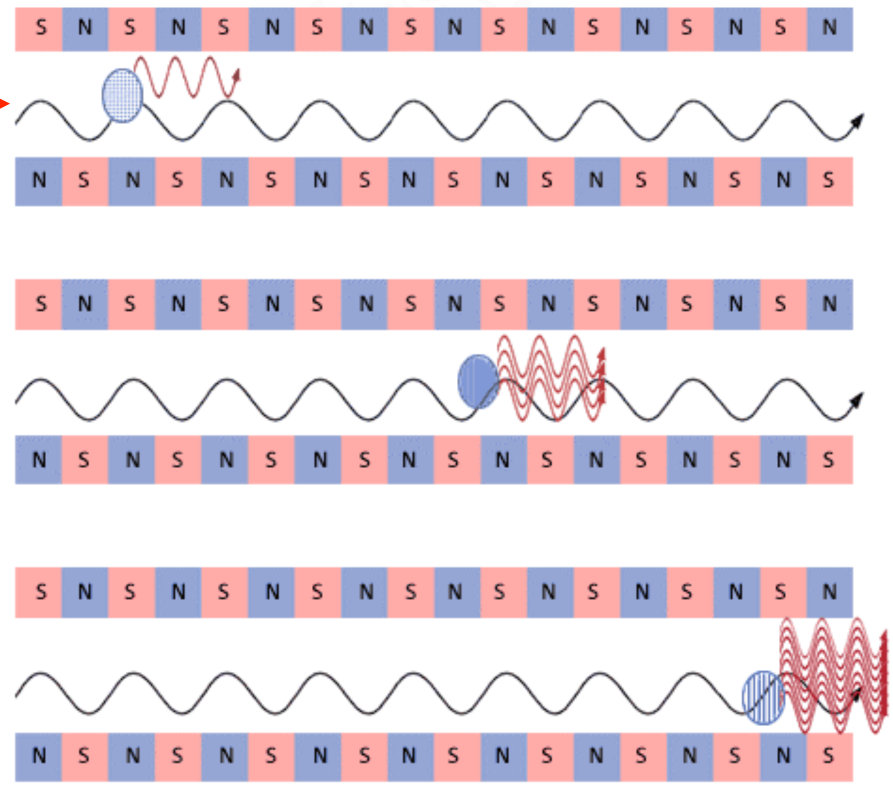
Peak current 2 kA

Undulator period 27 mm

FELs use Self Amplified Spontaneous Emission (SASE) to produce uniquely intense, ultrafast, coherent X-ray pulses



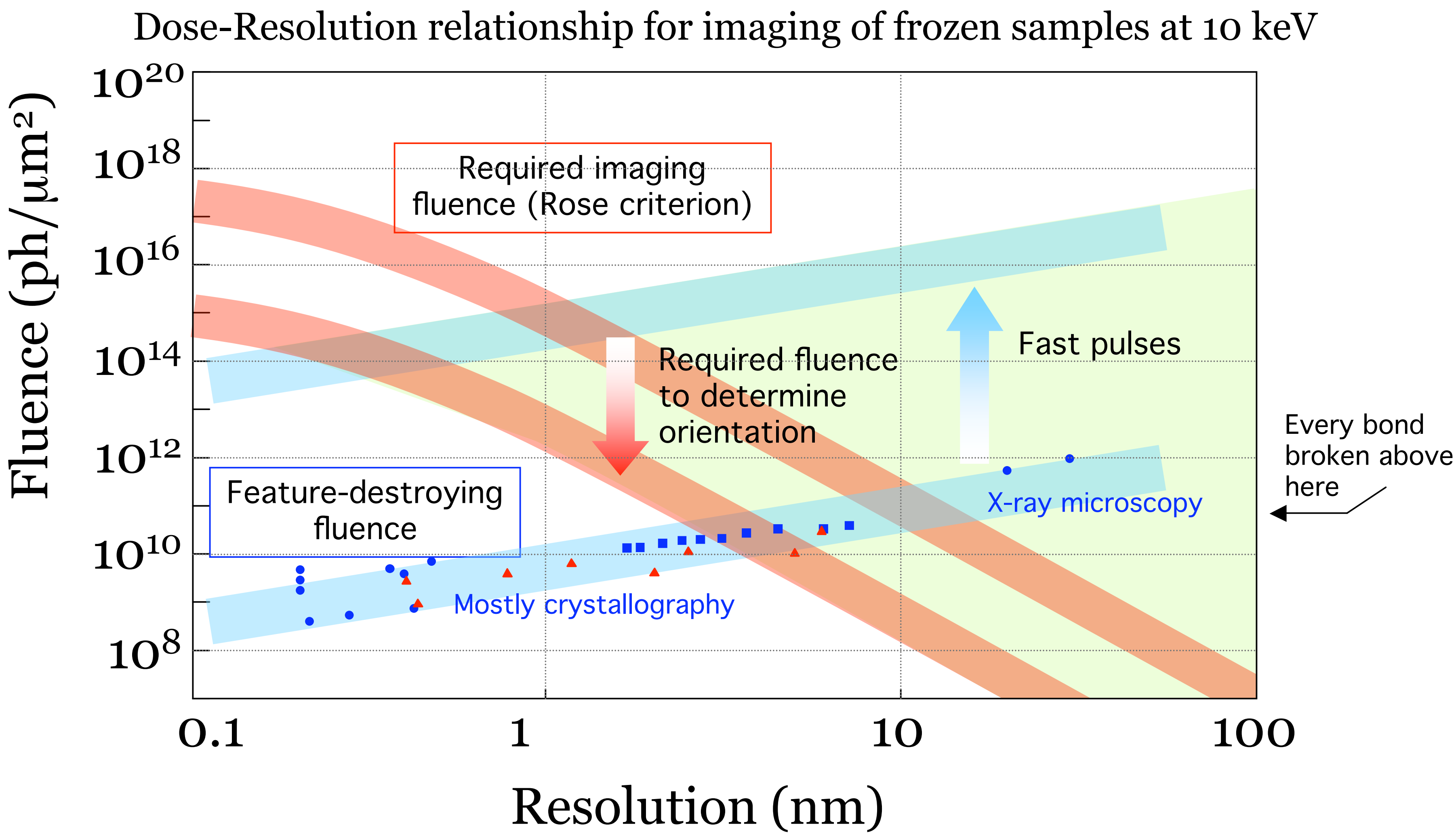
Electron bunch
(from LINAC)



X-ray pulses out
For FLASH:
λ = 6 - 32 nm
30 fs pulses
10^{12} photons
40 μJ (typical)
single mode

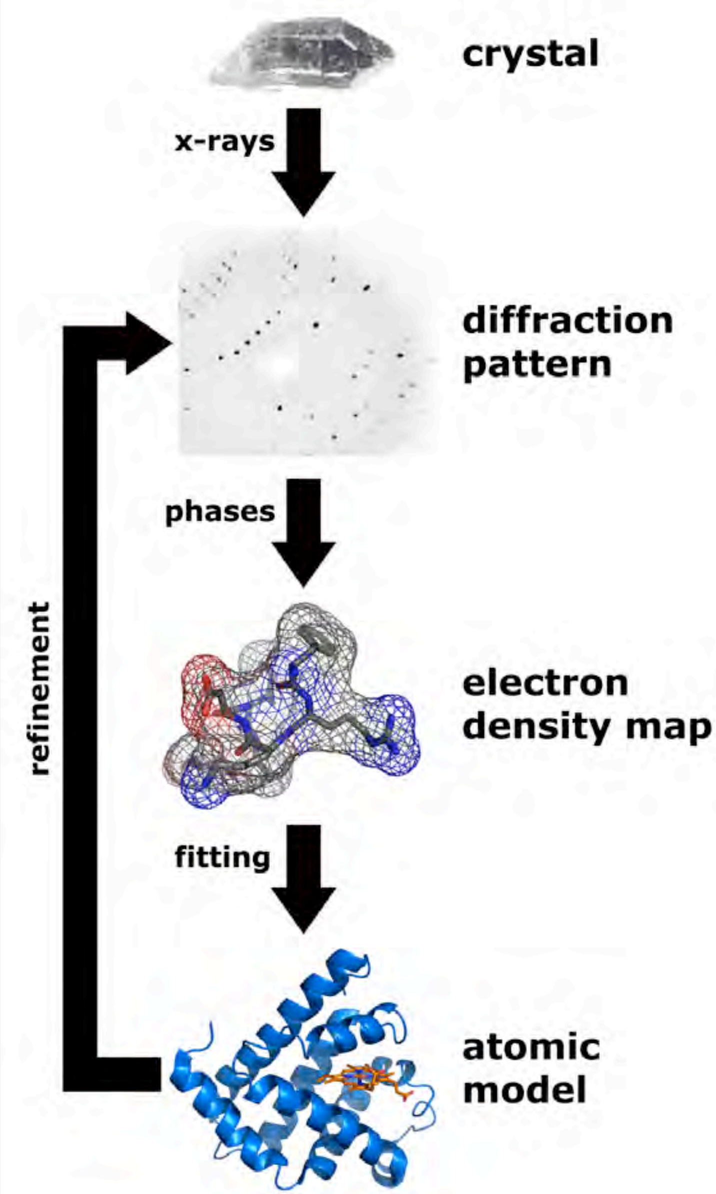
Graphics from:
vuv-fel.desy.de/
www-ssrl.slac.stanford.edu/lcls/

Imaging resolution is limited by radiation damage



Empirical data compiled by Malcolm Howells, LBL

Today, the majority of molecular structures are determined by x-ray crystallography

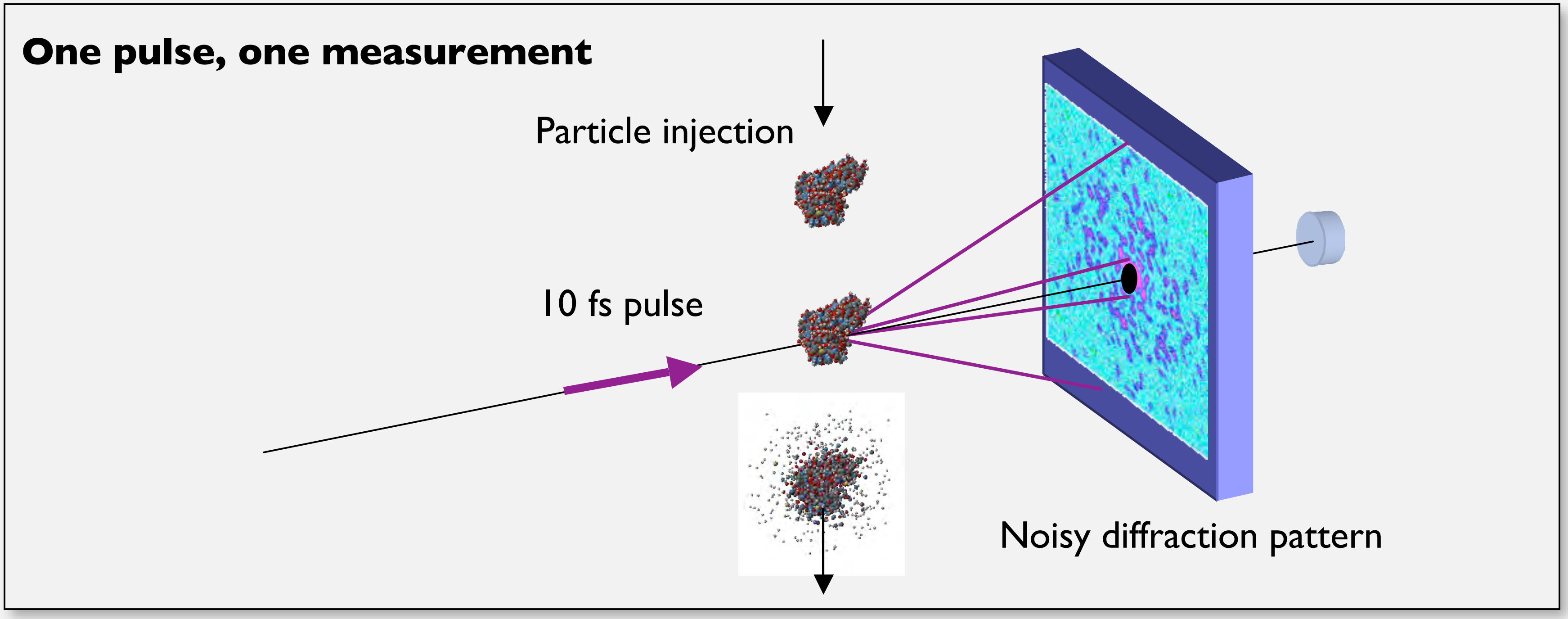


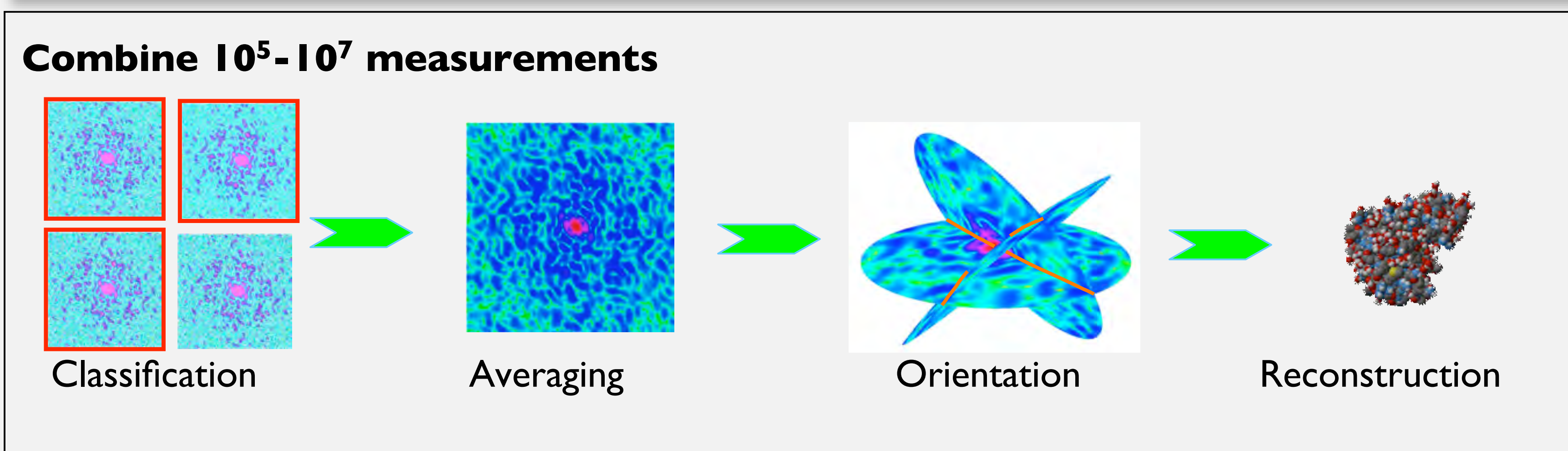
http://en.wikipedia.org/wiki/Image:X_ray_diffraction.png

- **Radiation damage is spread out over 10^{10} identical unit cells**
- **Diffraction from unit cells adds up coherently to form strong Bragg peaks**
- **> 40,000 structures solved (in protein data bank), but ~10,000 *distinct* structures**

- **The bottleneck is in growing crystals**

X-ray free-electron lasers may enable atomic-resolution imaging of non-crystalline biological macromolecules

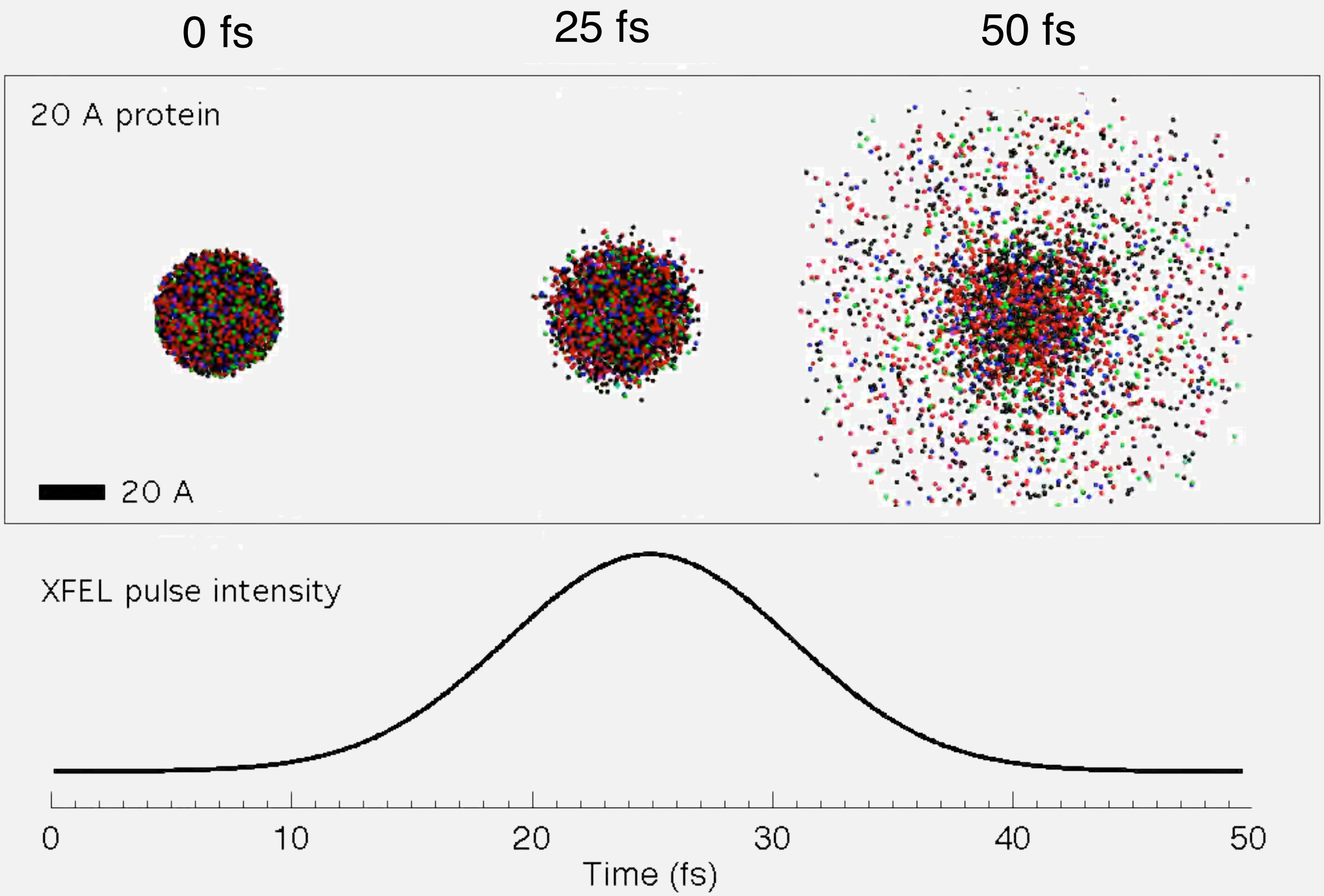




Destruction of the molecule is delayed by atom inertia: Fast pulses enable imaging before destruction

20 Å radius protein

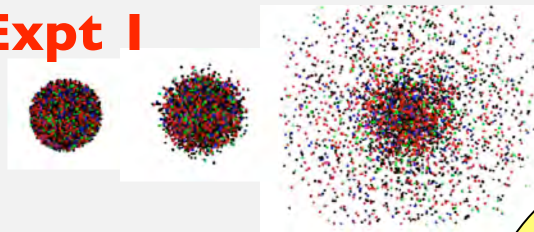
XFEL pulse intensity



Hydrodynamic model:
R. London, S. Hau-Riege, A. Szoke.

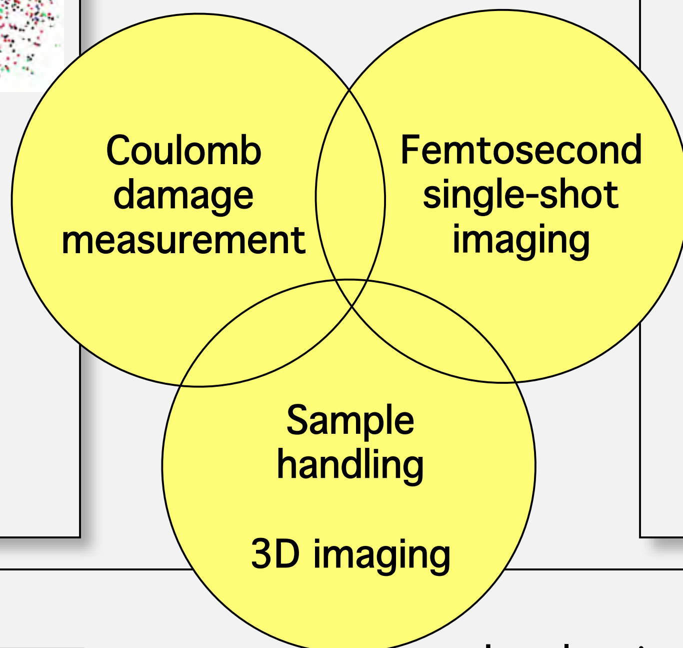
Technical challenge of working towards single molecule imaging is managed by focusing early work on key areas

Expt 1

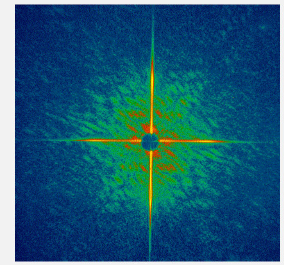


Experimental confirmation of rates of damage

Bulk samples and aerosols



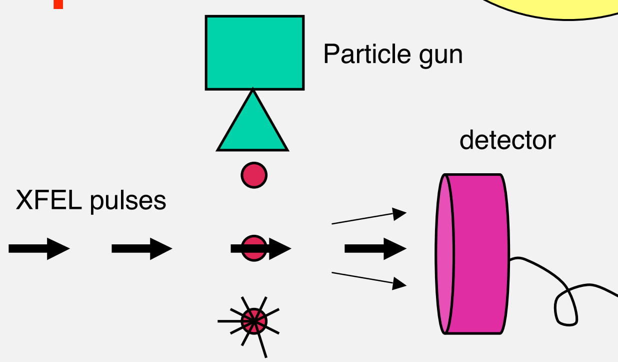
Expt 2



Experimental demonstration of imaging beyond the radiation damage limit

Test-structures and cells

Expt 3

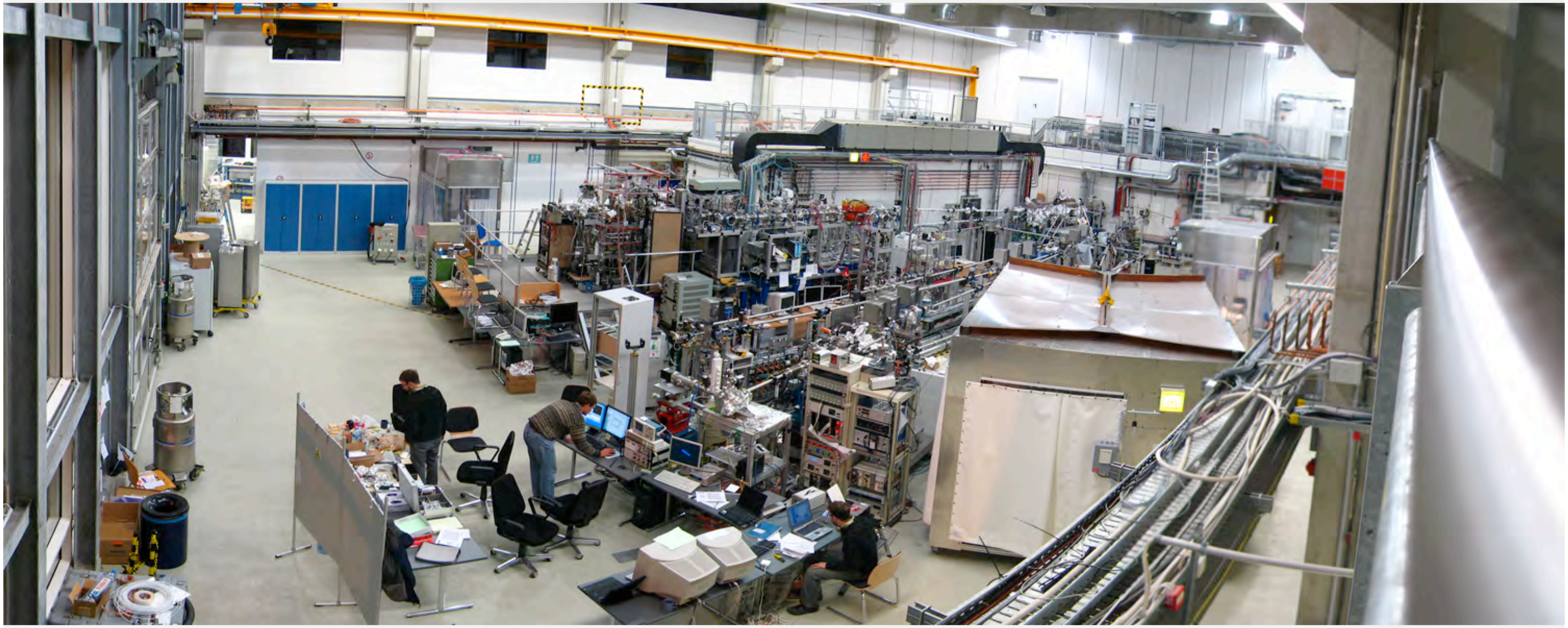


Lensless imaging of injected samples

3D diffraction imaging

Fixed samples at ALS
Injected samples at XFEL

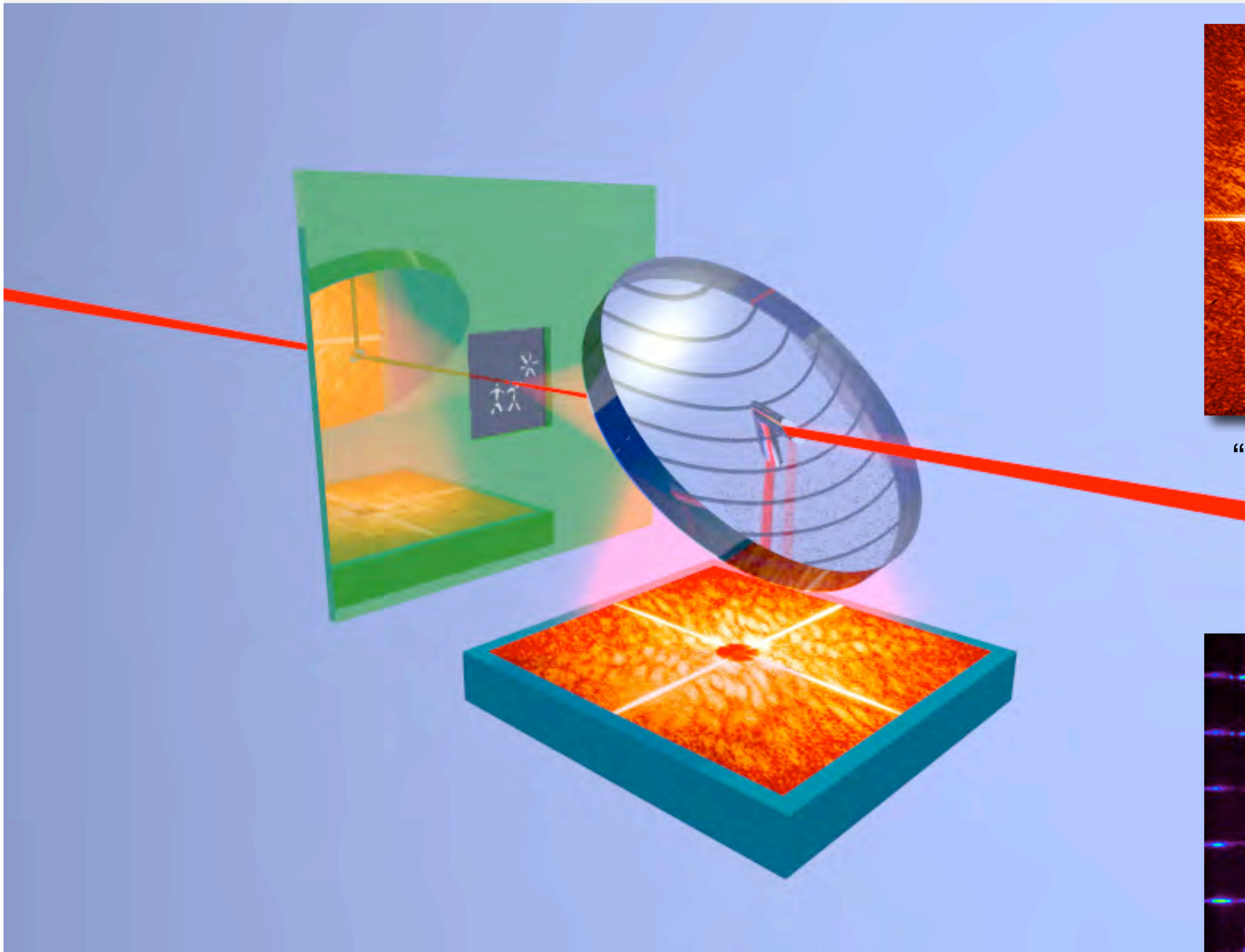
We have carried out experiments at the first operational soft-X-ray FEL in Hamburg

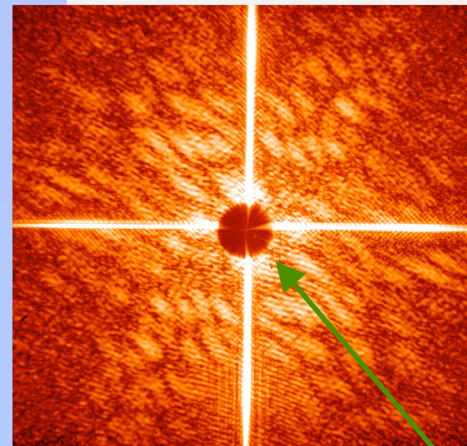


FLASH: The VUV-FEL at HASYLAB, DESY

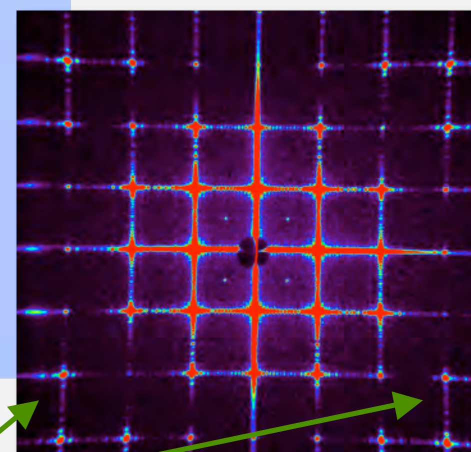
- User facility, FEL radiation to 6 nm wavelength (these experiments at 32nm)
- Initial FEL Operation August 2005 at 32 nm and <30 fs pulses, 10^{13} photons

Our diffraction camera can measure forward scattering close to the direct soft-X-ray FEL beam



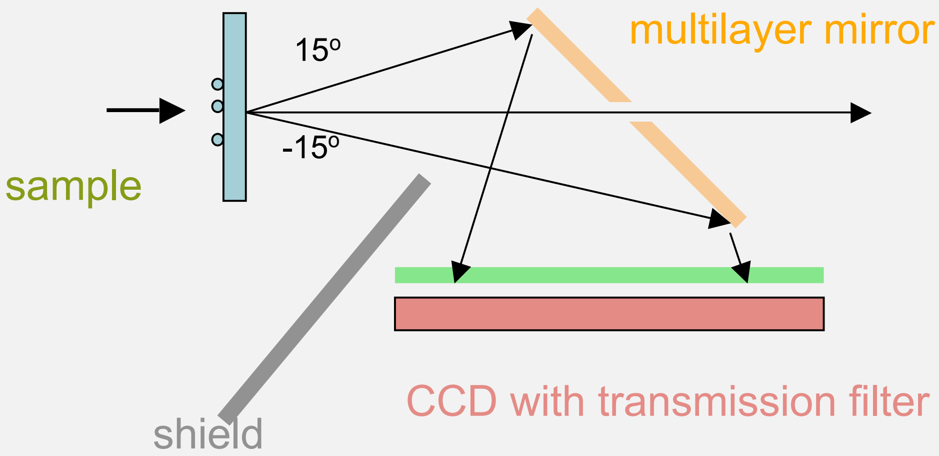


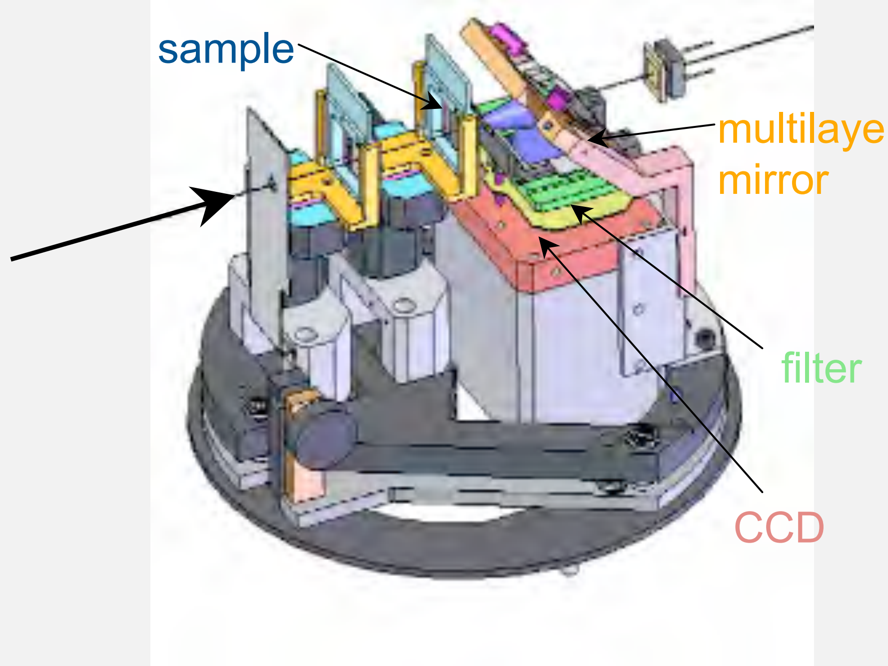
"Soft edge" prevents any
scatter from the hole

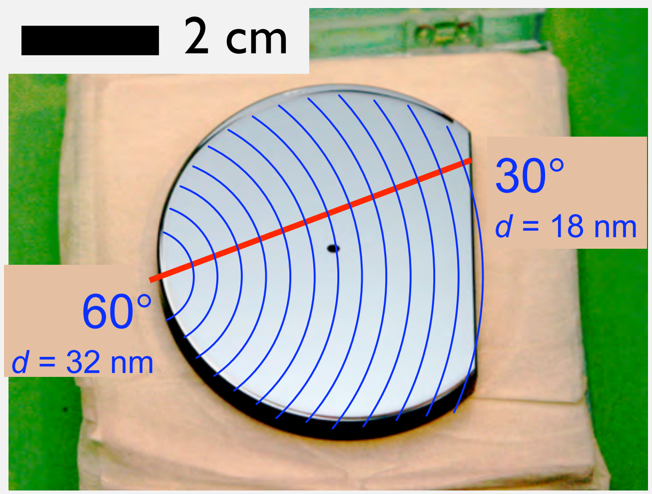


Multilayer reflectivity is uniform across
the 30° to 60° gradient

The VUV-FEL diffraction experiment is designed to measure forward scattering with high SNR





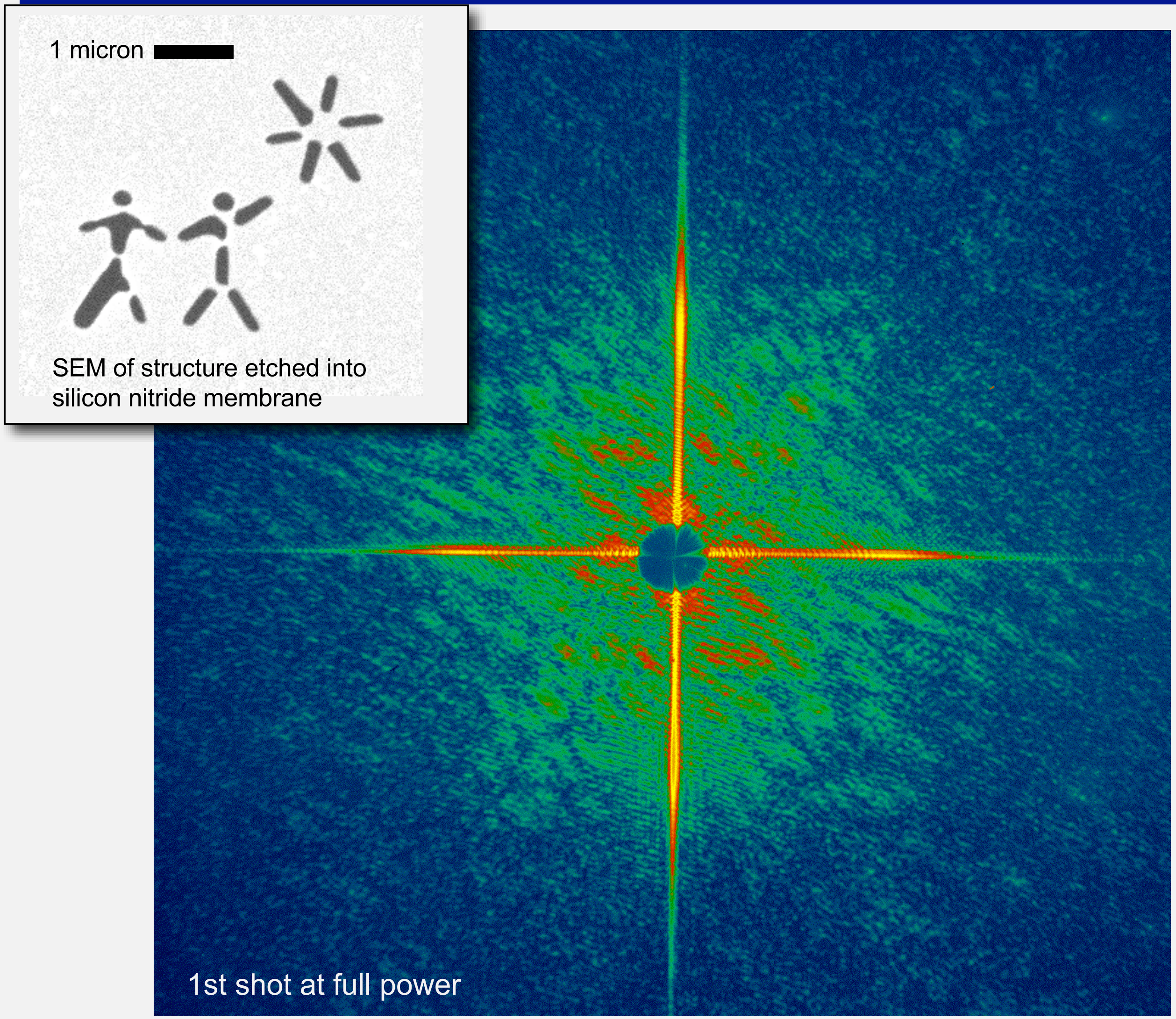


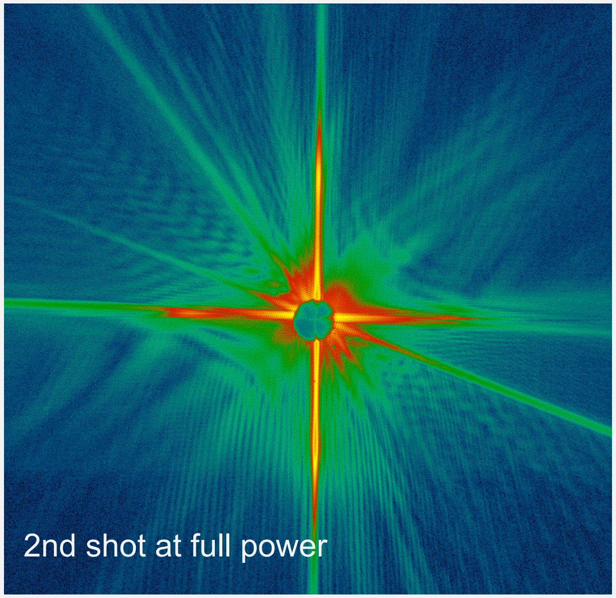
Sasa Bajt, Eberhard Spiller, and Jennifer Alameda

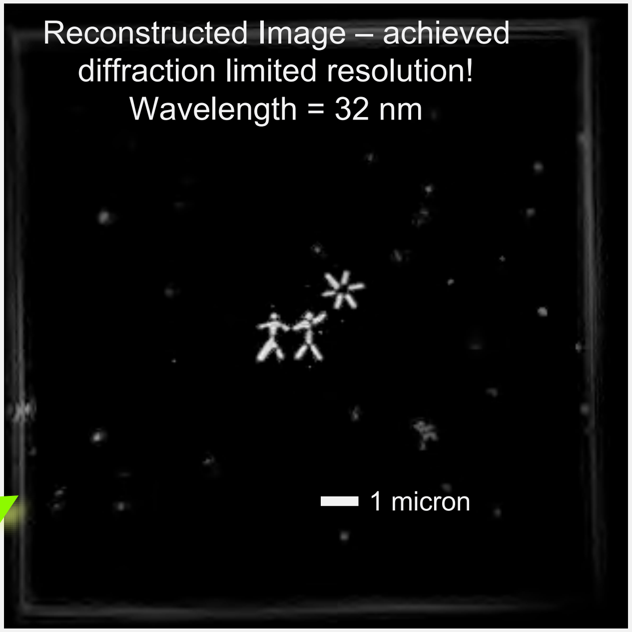
GRADED MULTILAYER MIRROR:
Si, Mo, and B_4C layers, period graded laterally. Variation matches angle of incidence (30° to 60°) to maintain Bragg condition for $\lambda = 32$ nm.
Reflectivity: 45% for 32 nm pulses.

The mirror protects the CCD and works as a
(i) bandpass filter (bandwidth = 9 nm at 45°)
(ii) filter for stray light (1% off-axis reflectivity)
(iii) low-scatter beam-stop

We have successfully reconstructed images from ultrafast FEL diffraction patterns



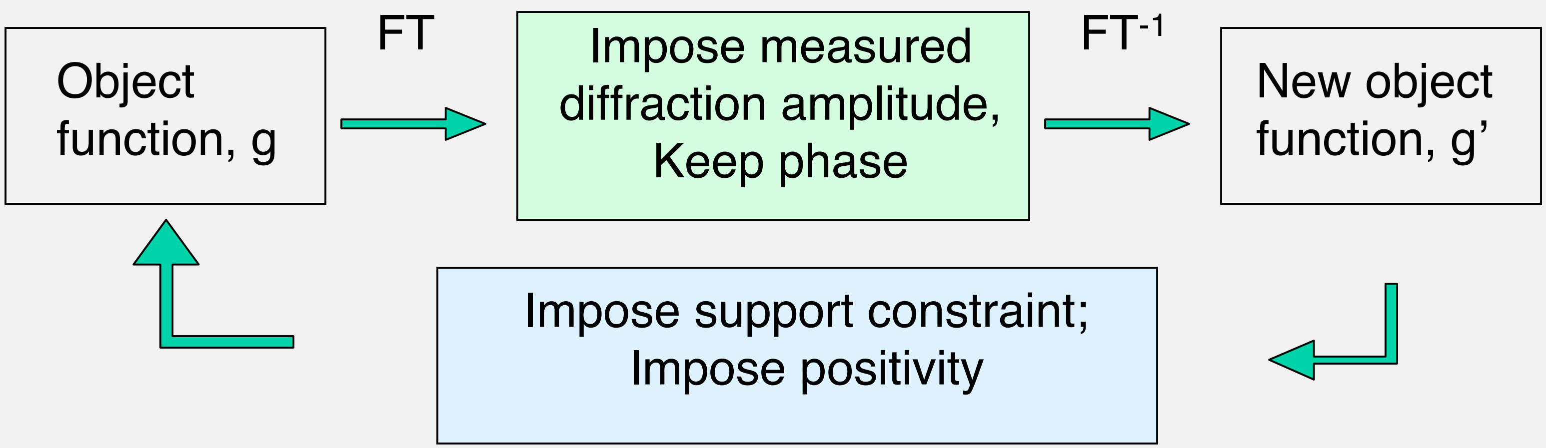


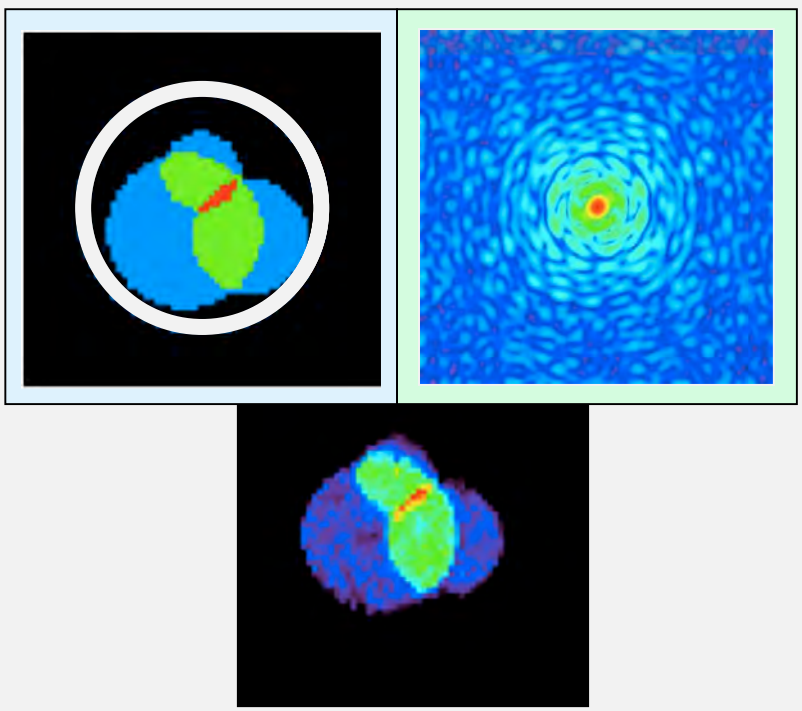


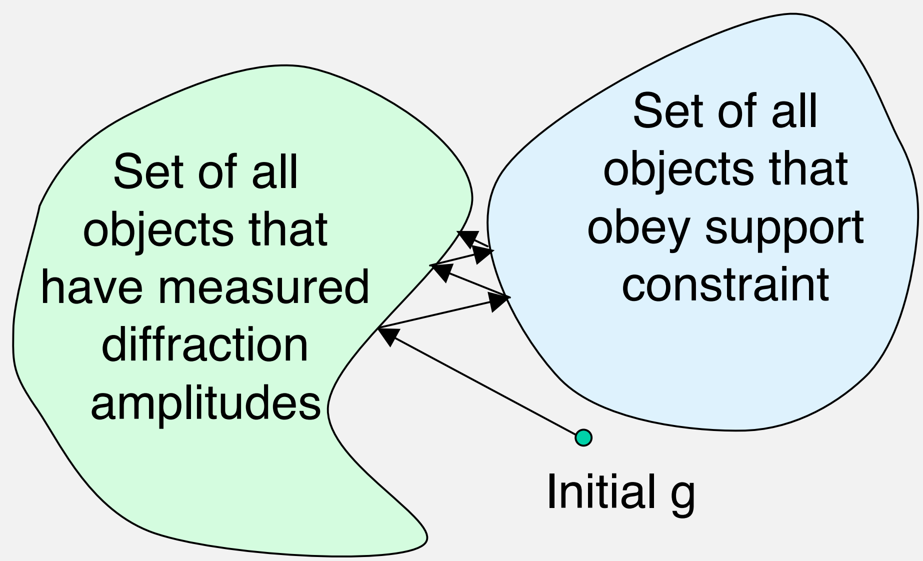
Pulse energy: 10 μJ or 1 J/cm²
Dose in Si_3N_4: 10^5 J/g or 22 eV/atom
Temperature of 6×10^4 K, or 5.2 eV, ionization/atom of 2.5
Surfaces will expand at sound speed: 1.3×10^6 cm/s
In 25 fs, material will not move more than 3 Å

Edge of membrane support also reconstructed

Images are reconstructed using iterative transform phase retrieval algorithms

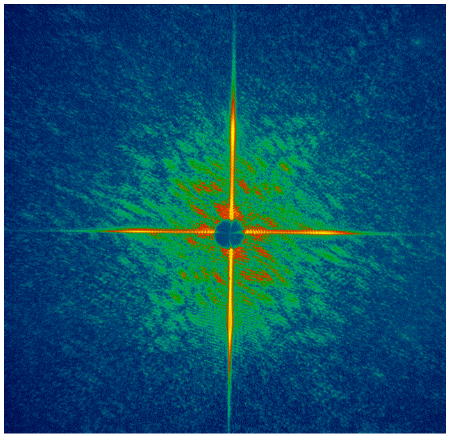




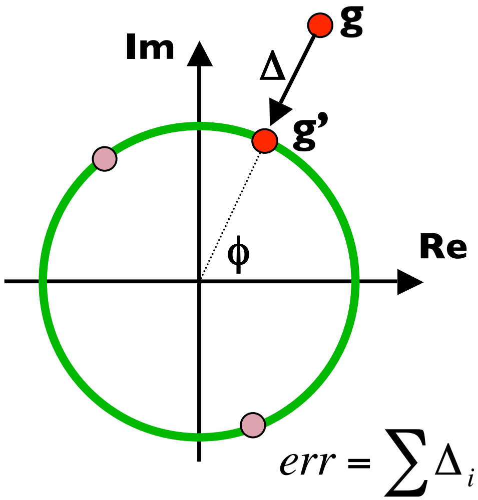


Imaging can be accomplished using iterative transform phase retrieval algorithms

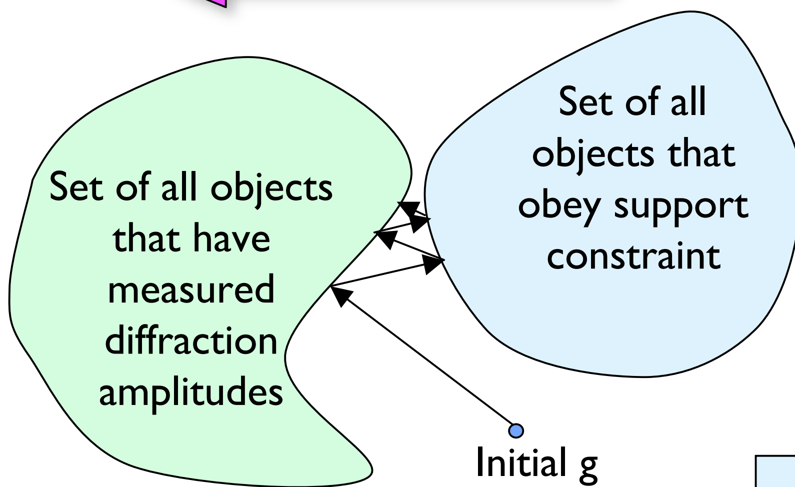
Diffraction space



Modulus projection
$P_{mod}(\mathbf{g}_i)$



FT



Periodically update object support (shrinkwrap)

FT^{-1}

Repeat {

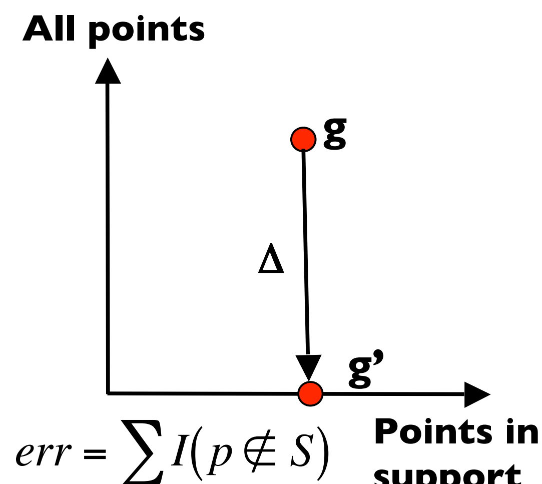
…

…

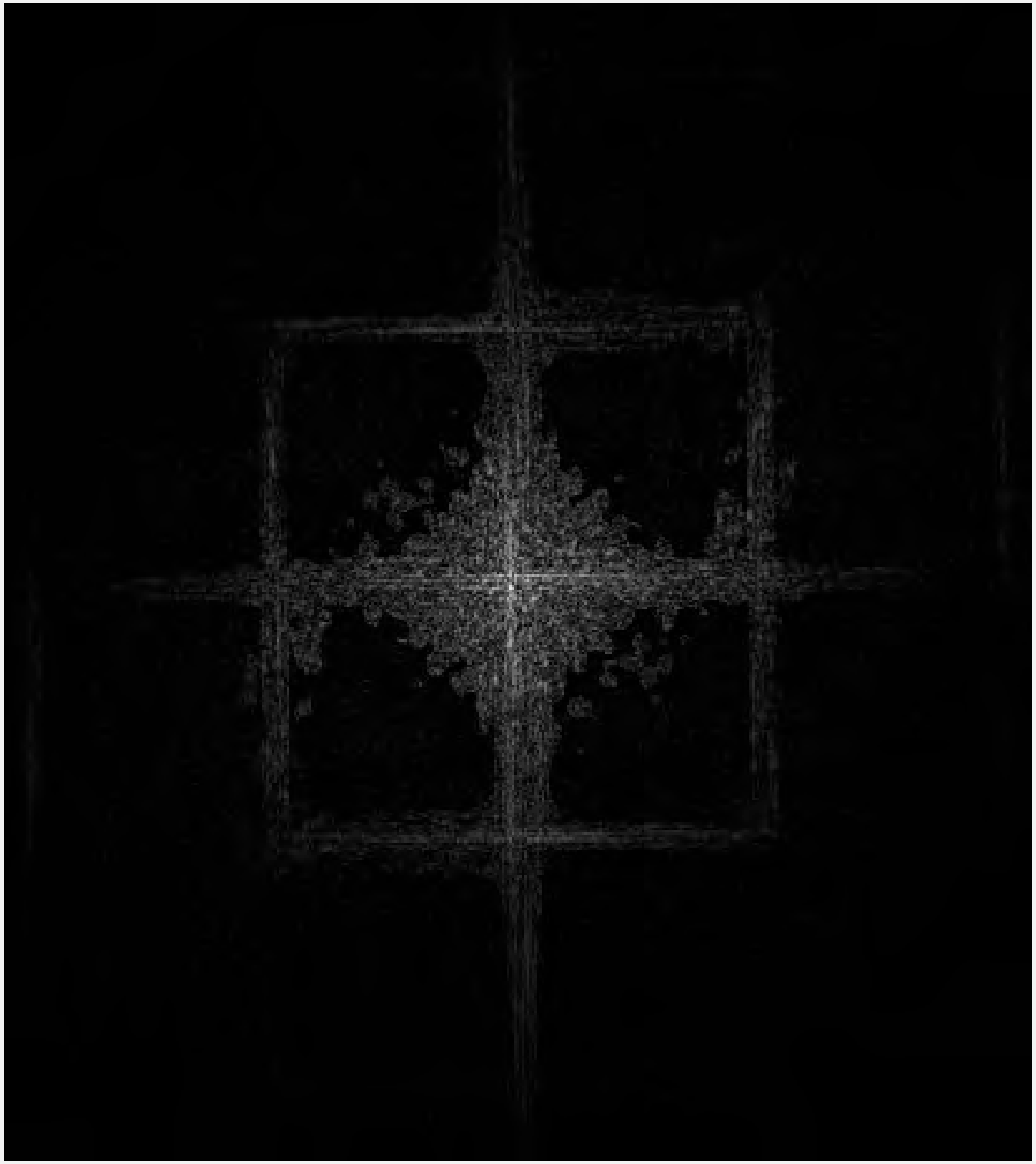
}
until constraints satisfied
(error is small)

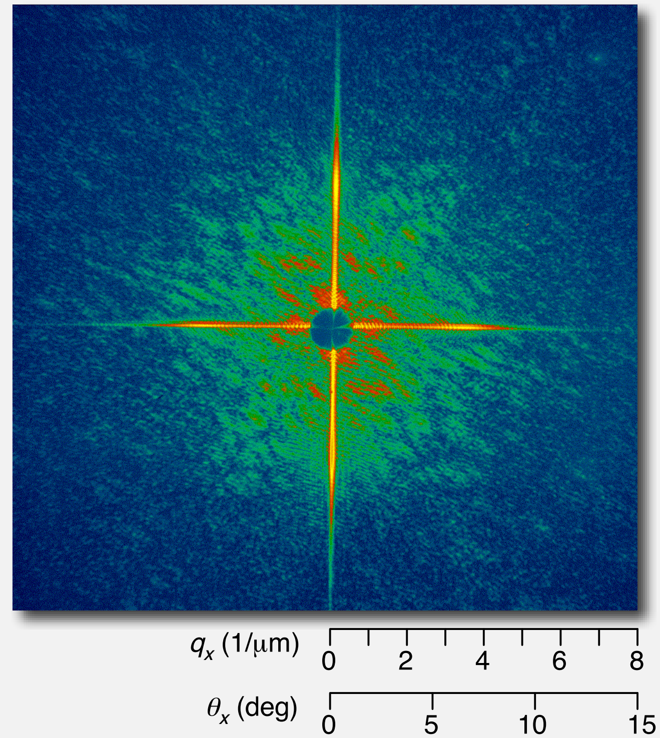
Real space

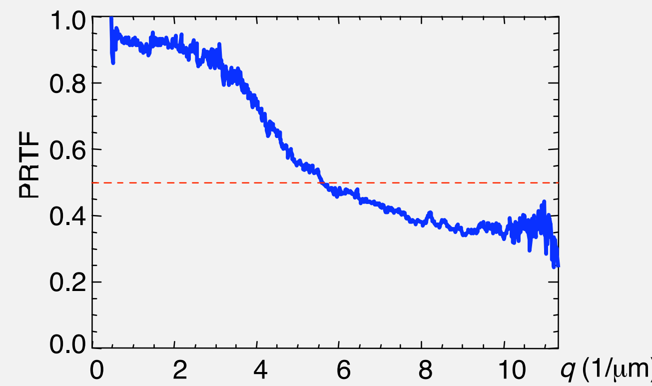
eg: Support projection
$P_s(g_i)$



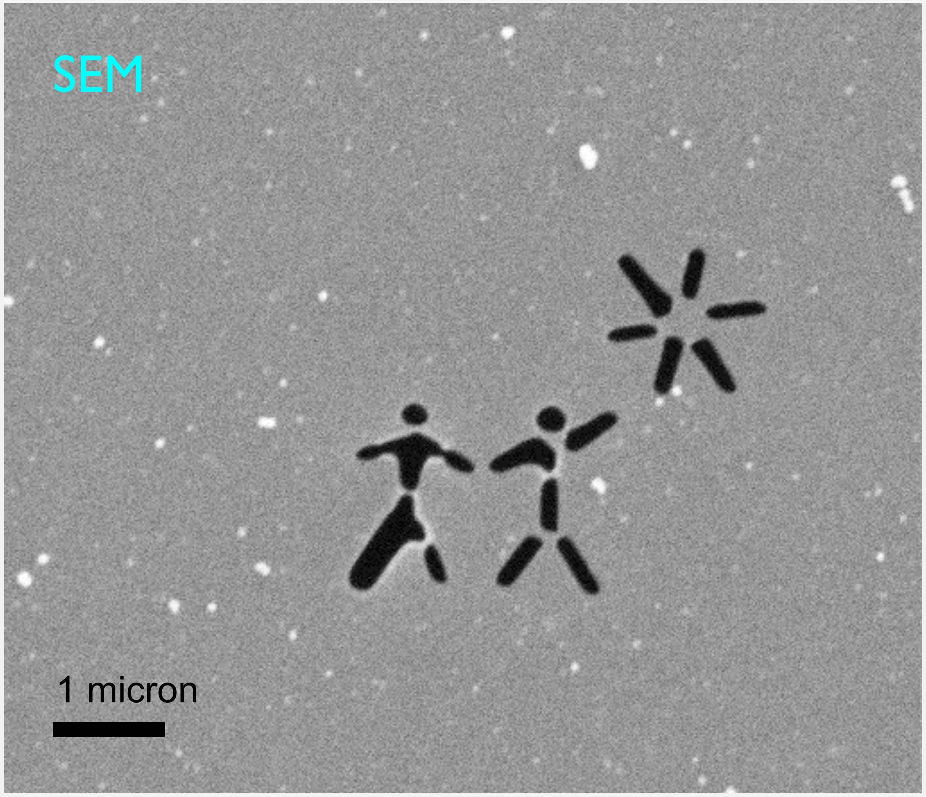
We perform ab initio image reconstruction with our "Shrinkwrap" algorithm

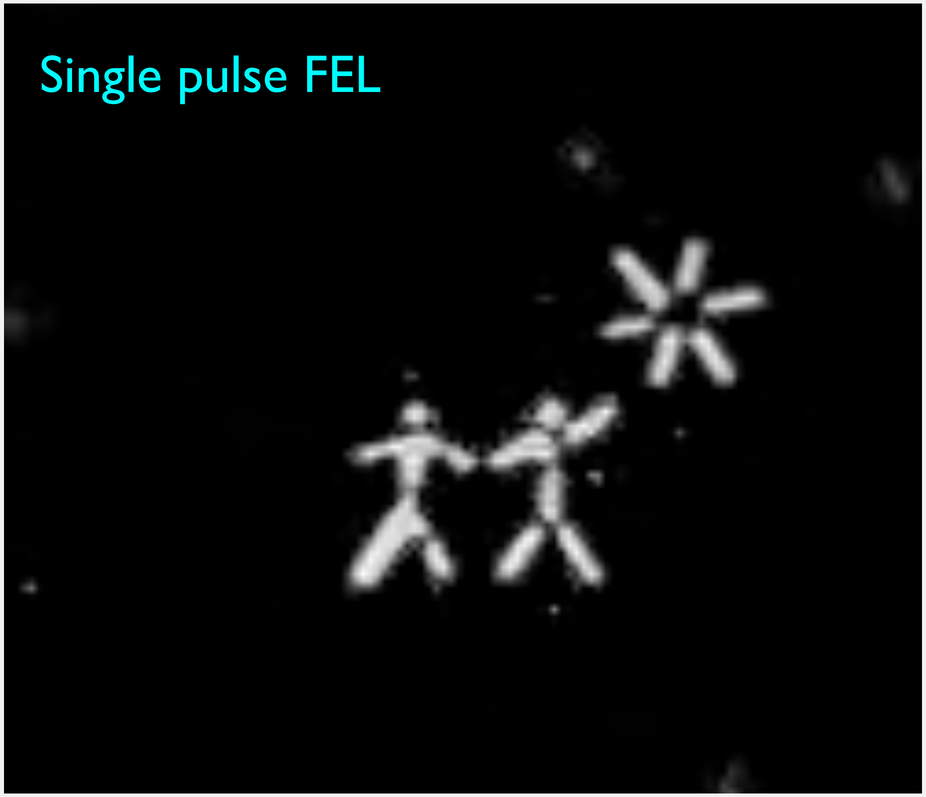




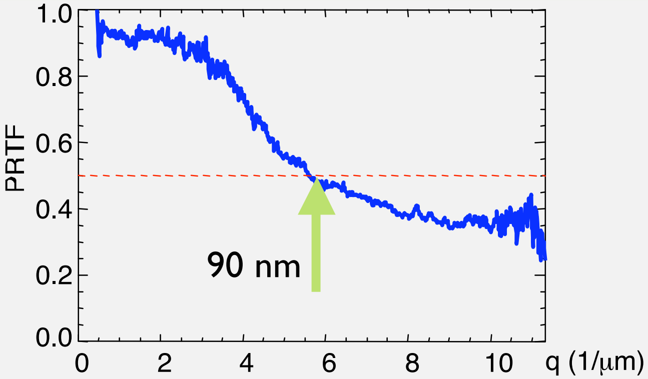


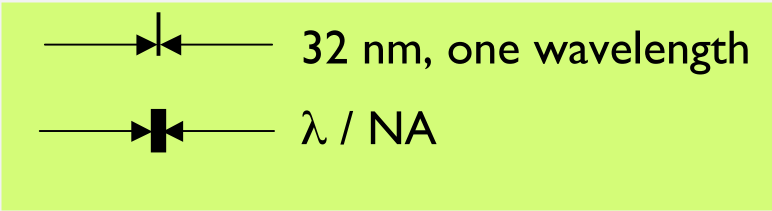
The reconstruction is carried out to the diffraction limit of the 0.26 NA detector



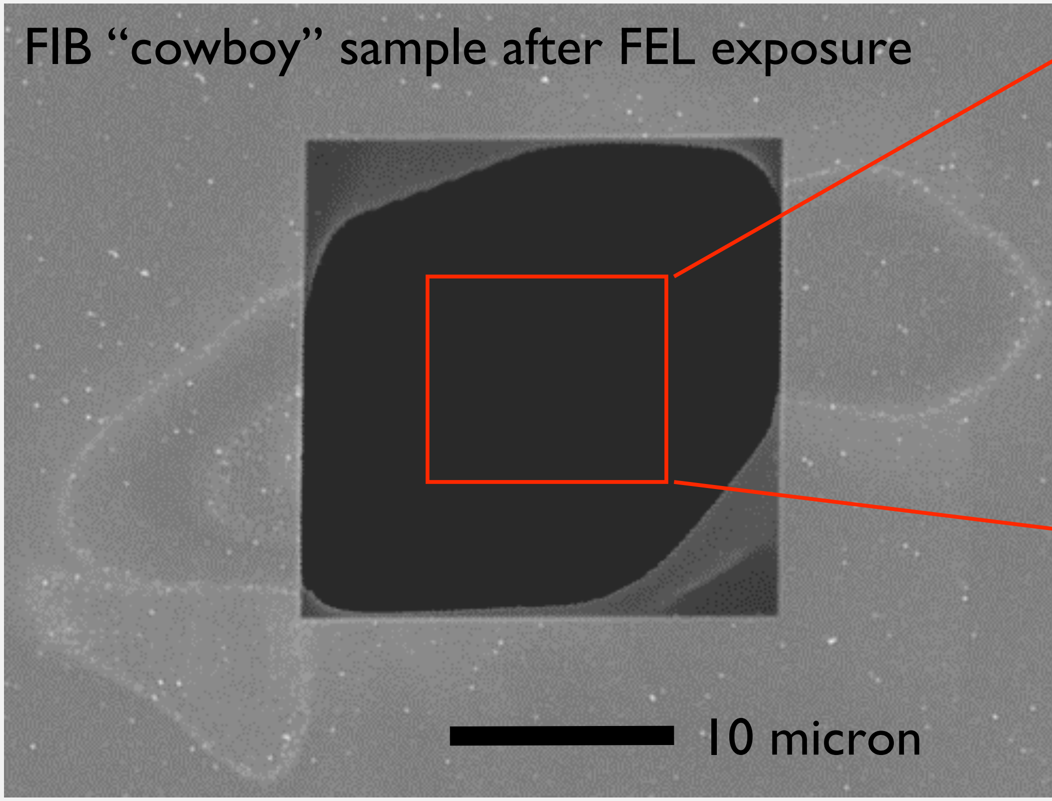


Phase-retrieval transfer function gives an estimate of the resolution of the reconstructed image

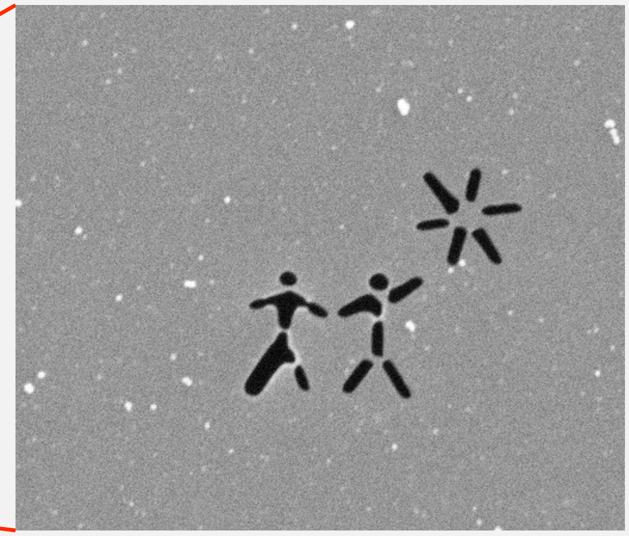


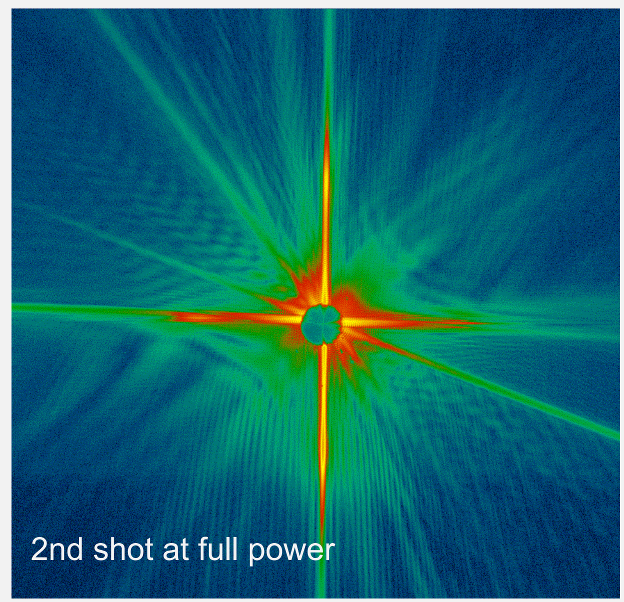


The sample is significantly damaged by the FEL pulses



What the 1st shot saw…





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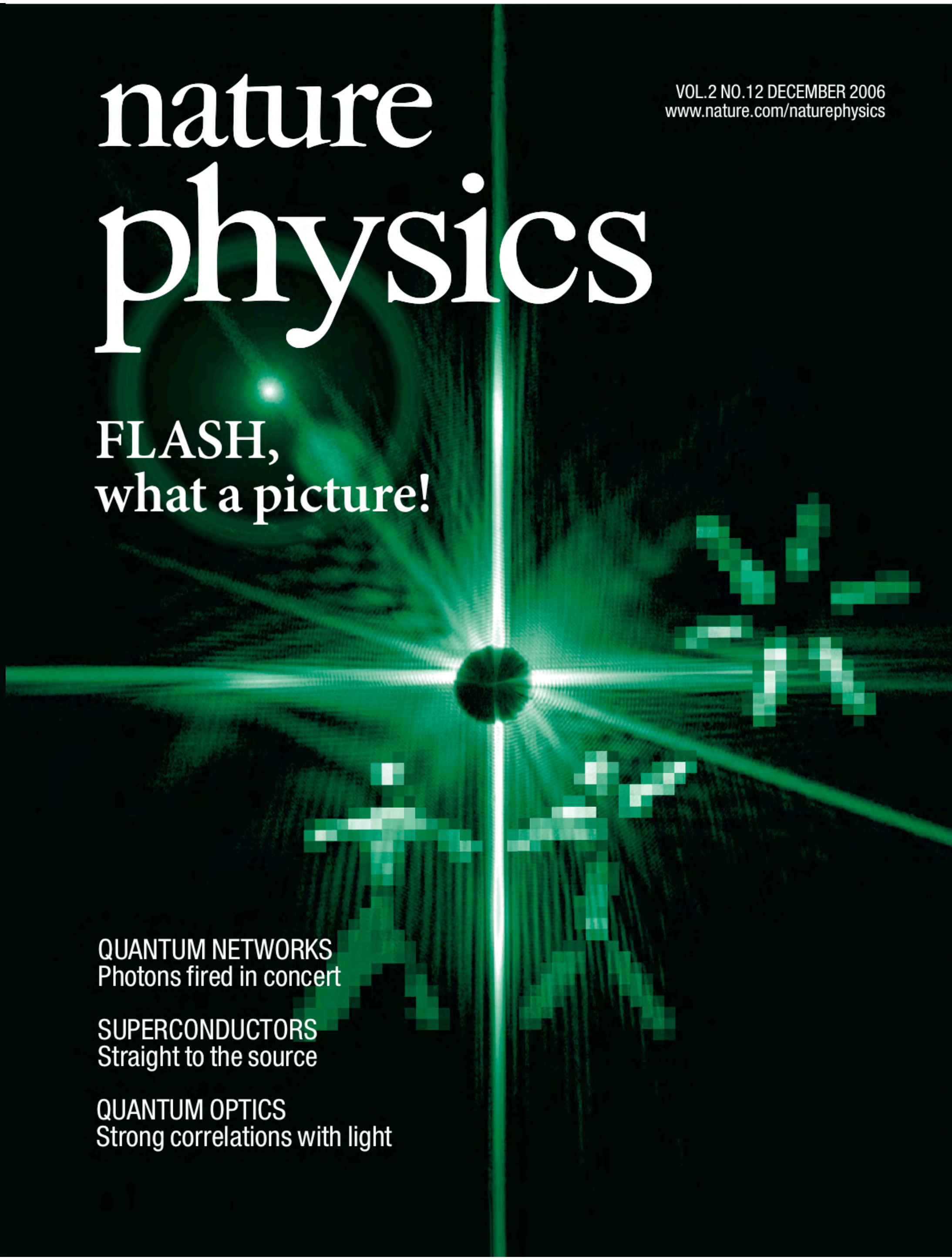


nature physics

VOL.2 NO.12 DECEMBER 2006
www.nature.com/naturephysics

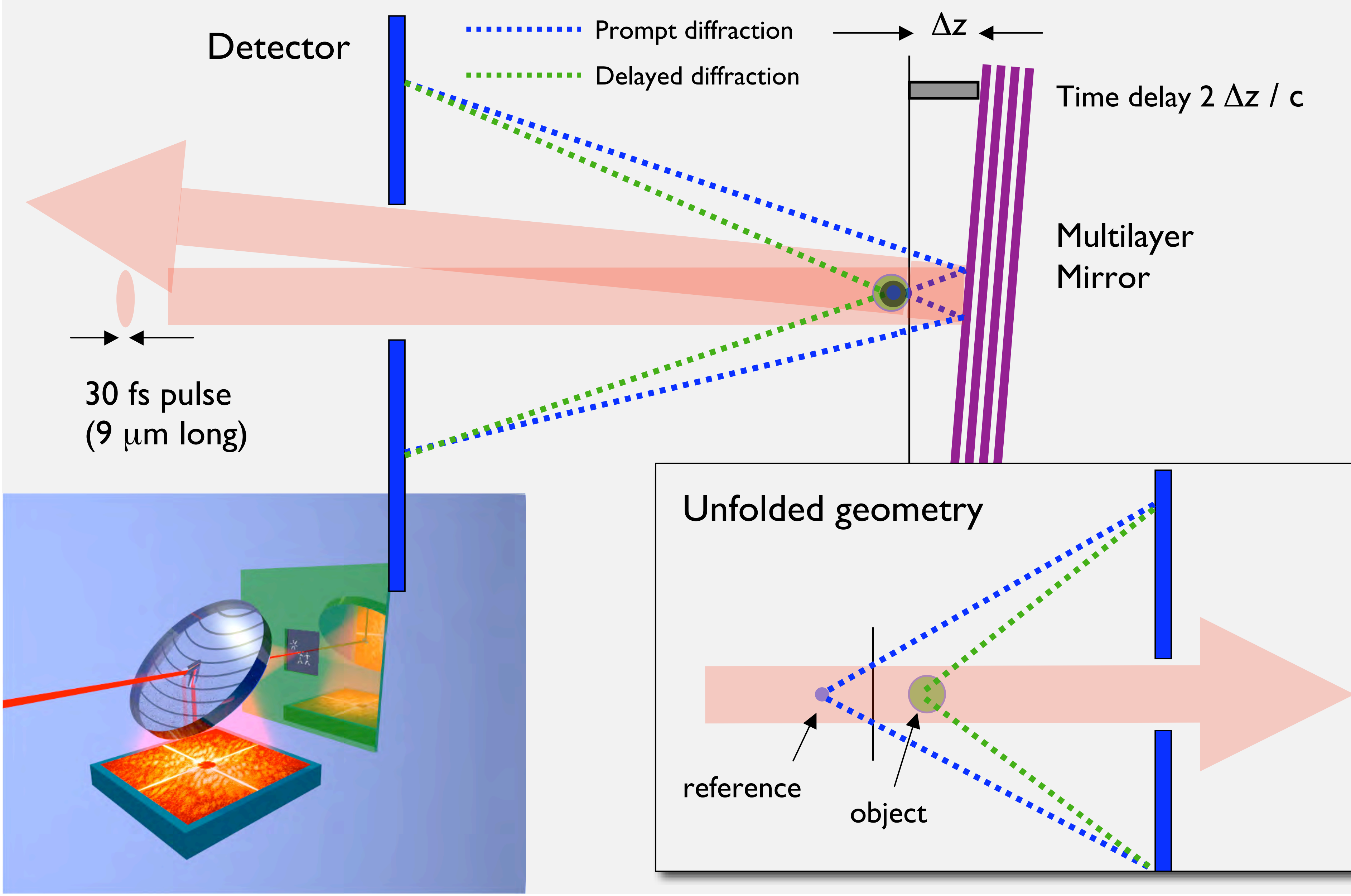
FLASH, what a picture!

QUANTUM NETWORKS
Photons fired in concert

SUPERCONDUCTORS
Straight to the source

QUANTUM OPTICS
Strong correlations with light

We invented a new method called femtosecond time-delay holography



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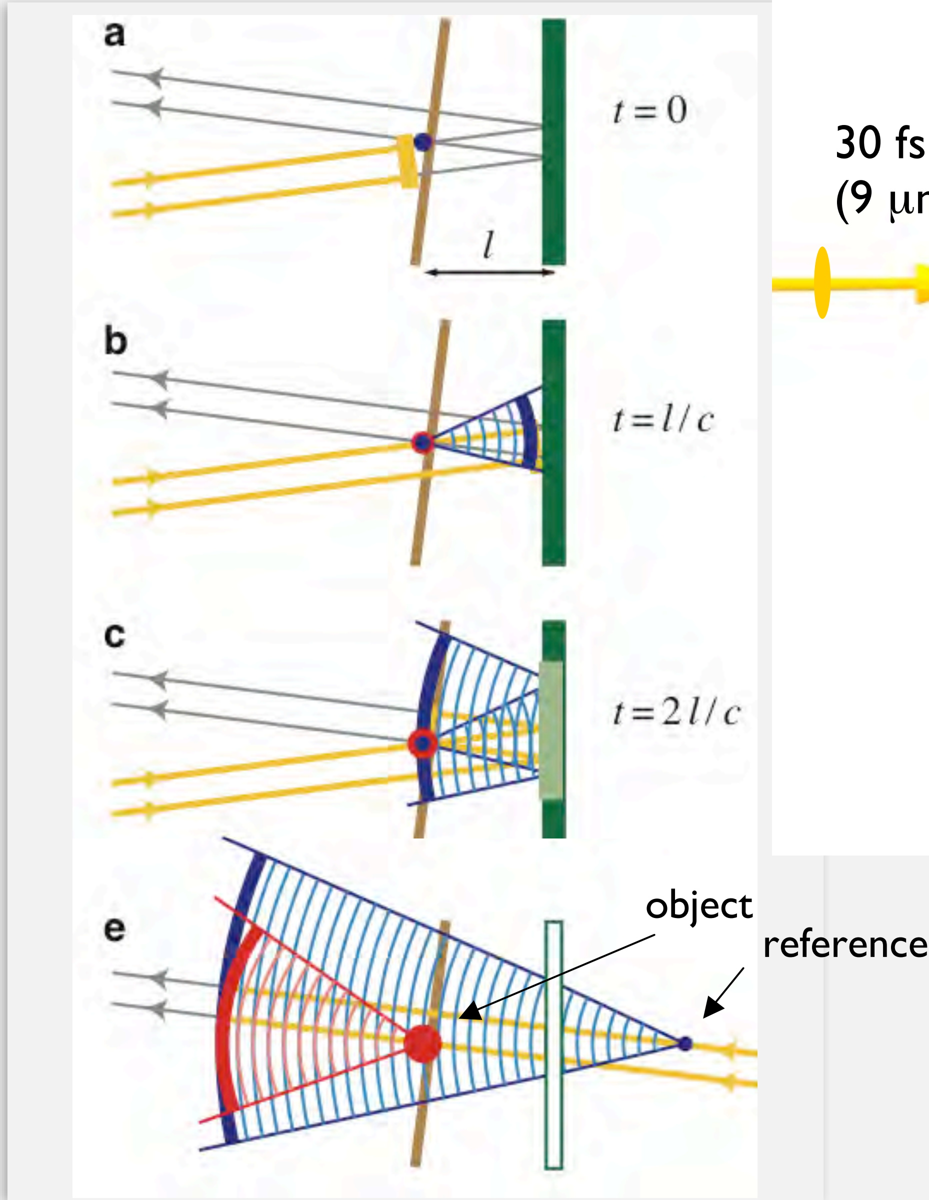
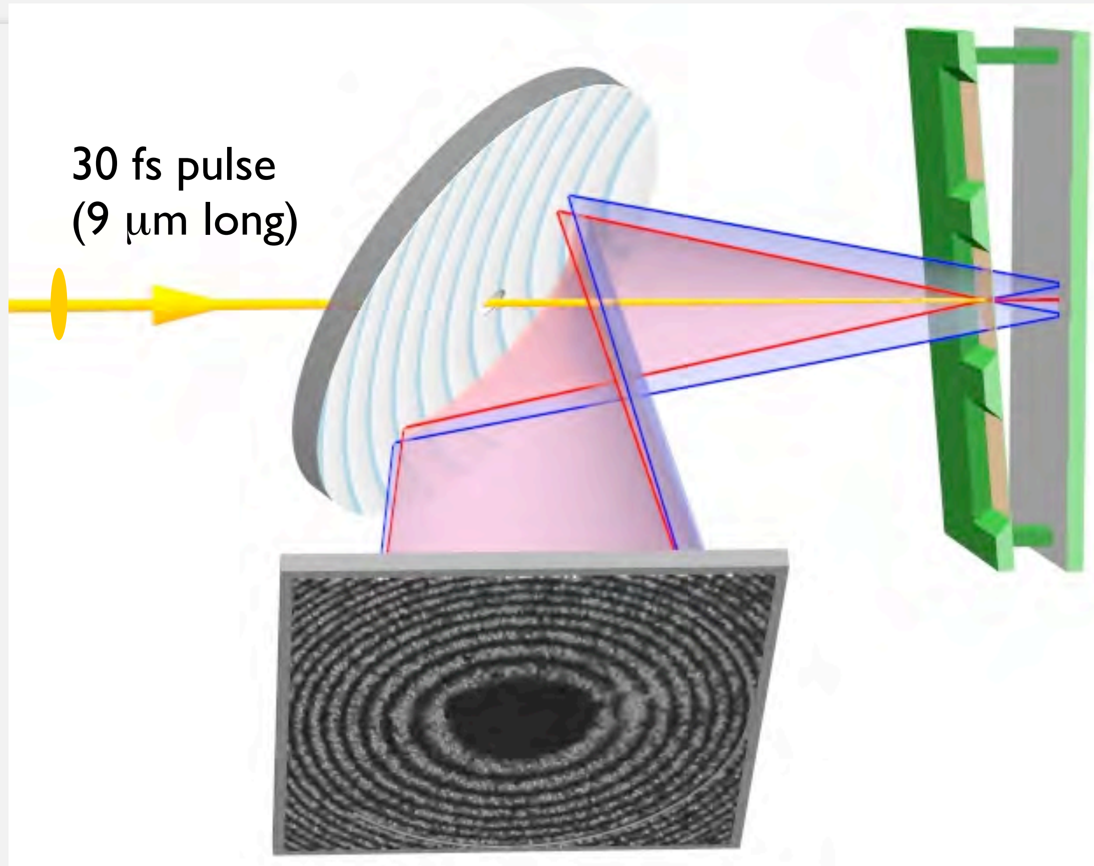


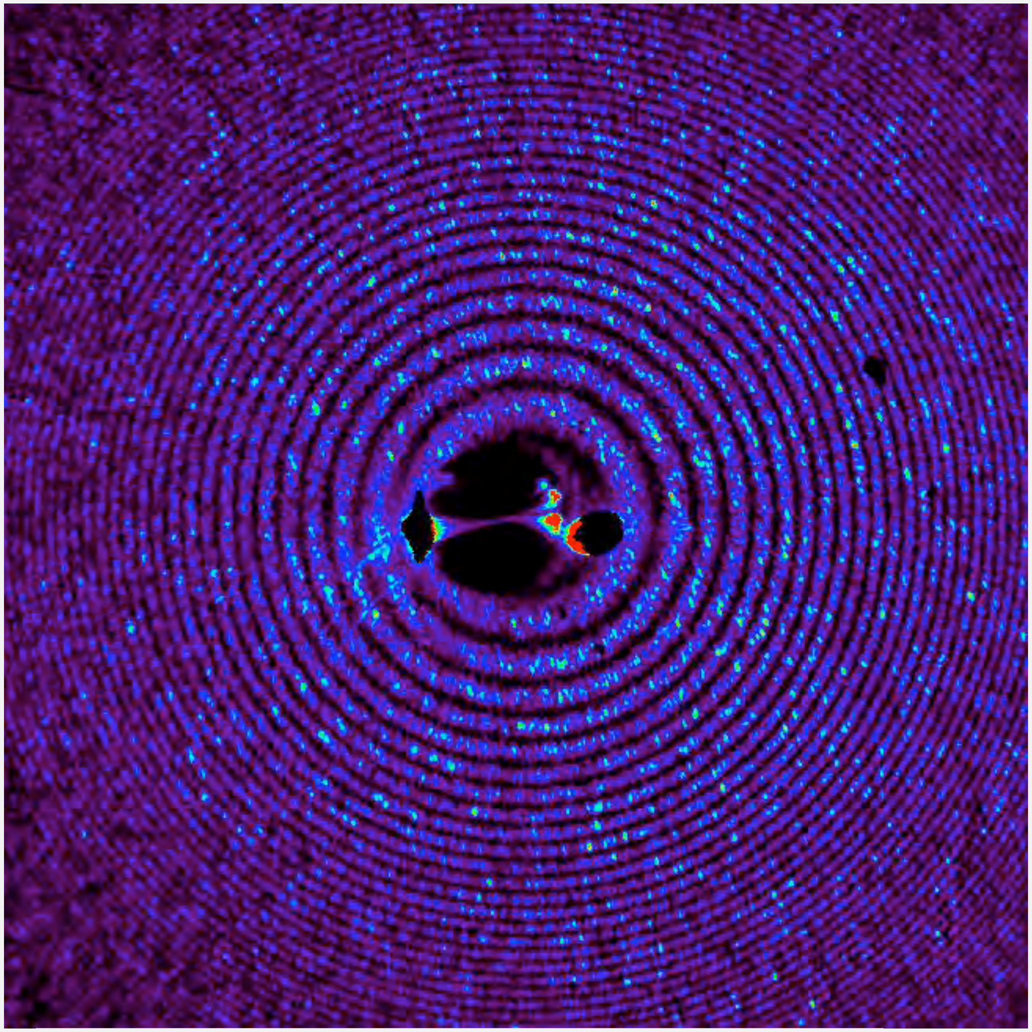


Prompt diffraction

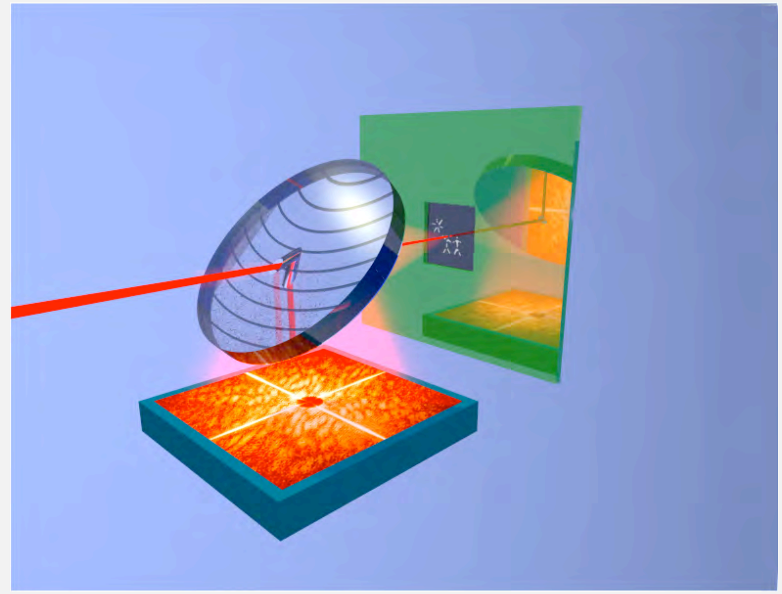
Delayed diffraction

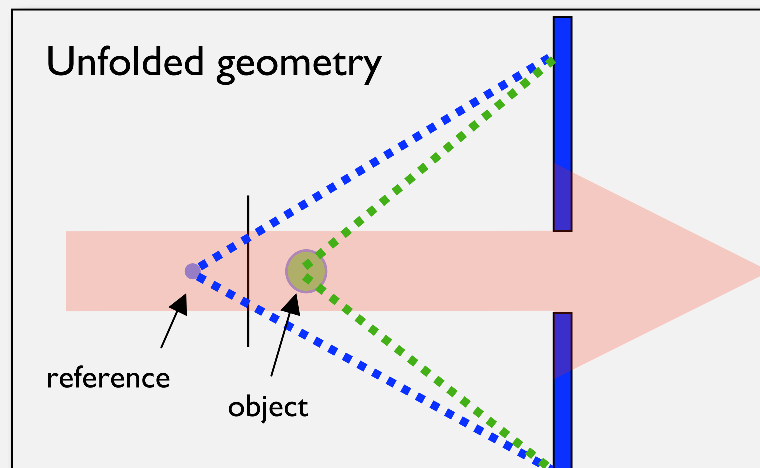
Time delay $2\ l\ /\ c$

FLASH results: Femtosecond time-delay holography with 30 fs time resolution



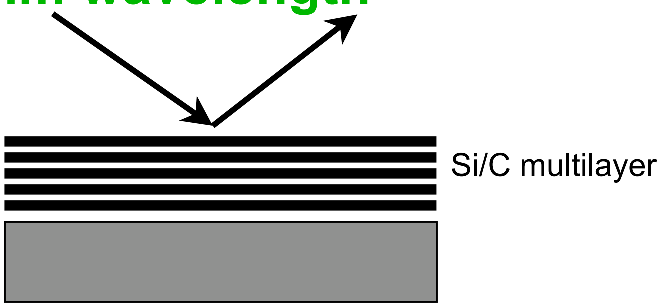
Single shot ultrafast time-delay X-ray hologram, with 300 fs delay



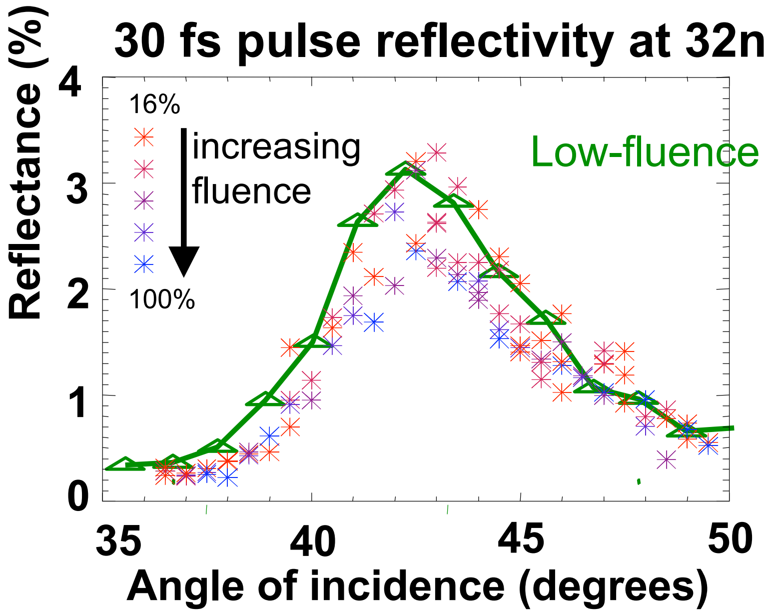


Experiments at FLASH demonstrate that sample structure to 0.3 nm can be obtained before sample destruction

During 30 fs pulse (10^{14} W cm^{-2})
32 nm wavelength

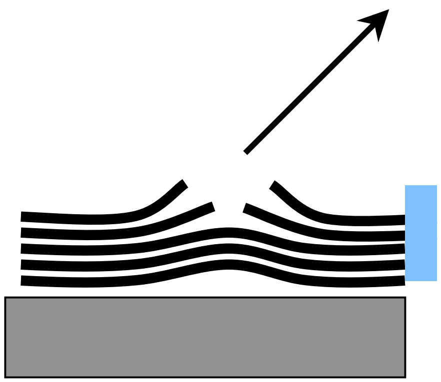


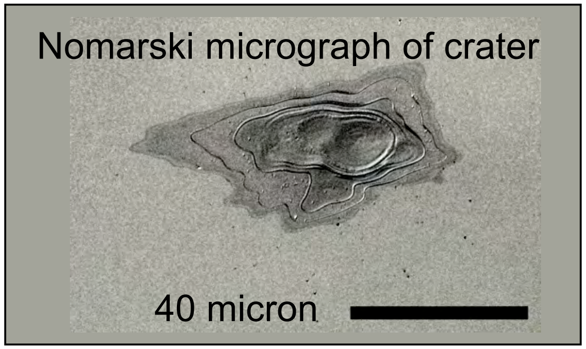
Reflectivity unchanged
Multilayer *d* spacing not changed by more than 0.3 nm

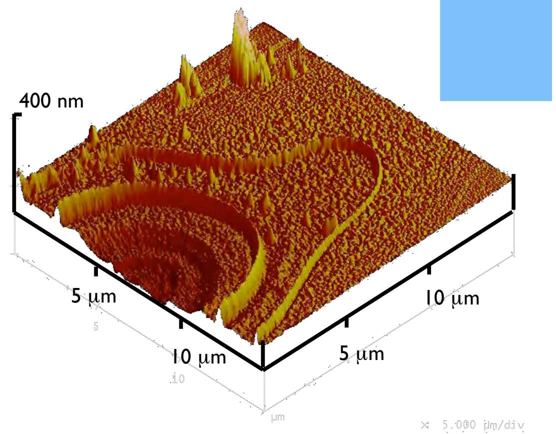


After pulse

Plasma forms, layers ablate



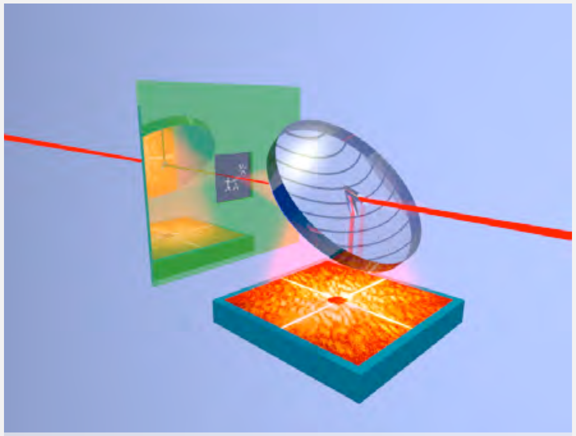




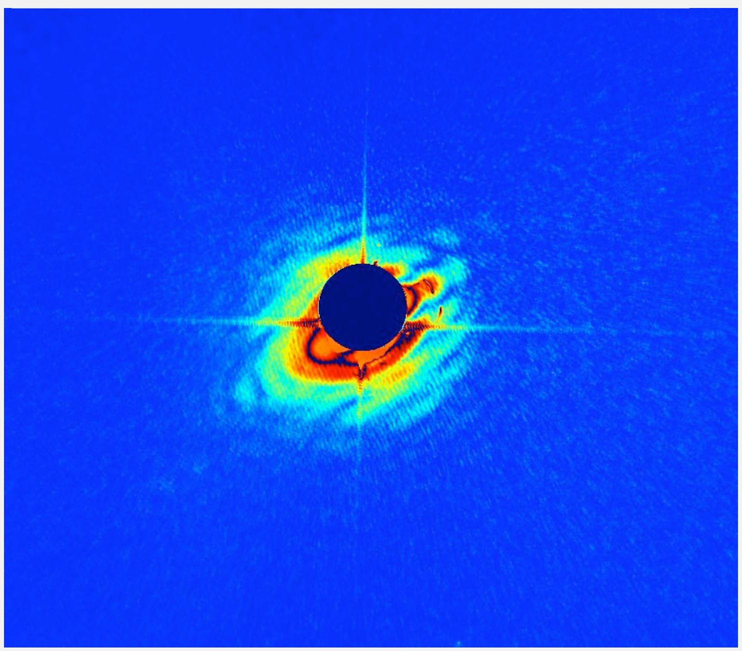
J. Kryzwinski, R. Sobierajski, H. Chapman et al

Recent results from FLASH: Biological structures

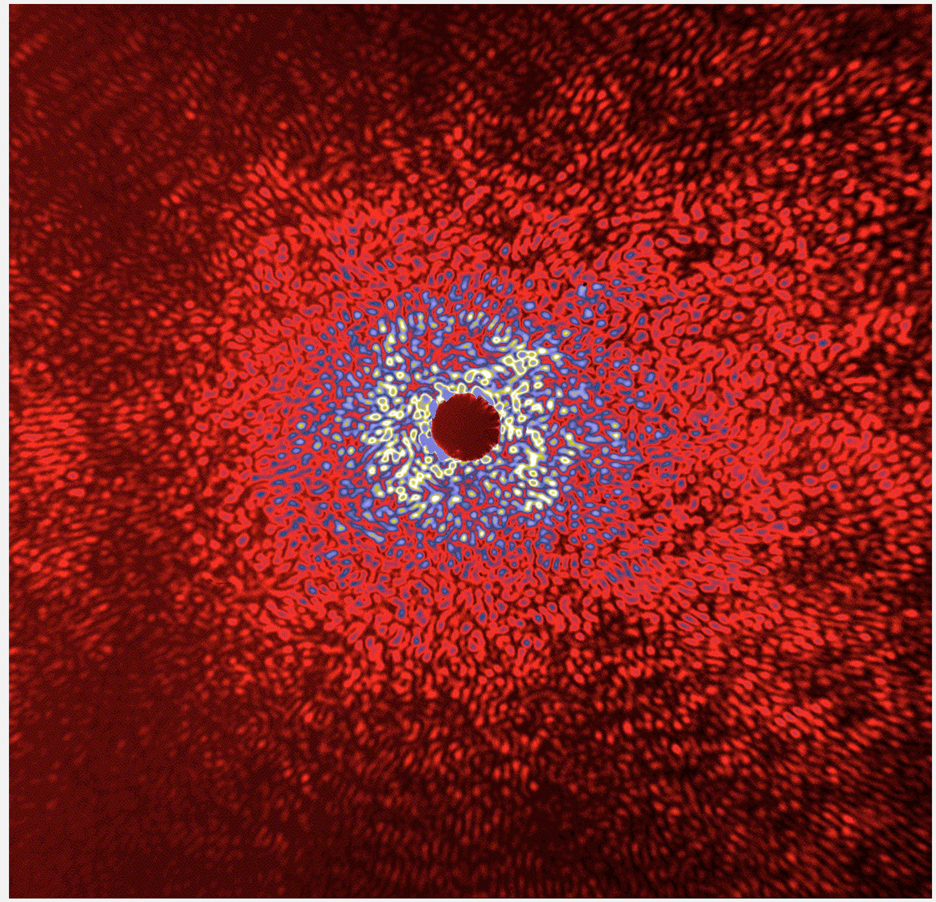
Forward scattering geometry



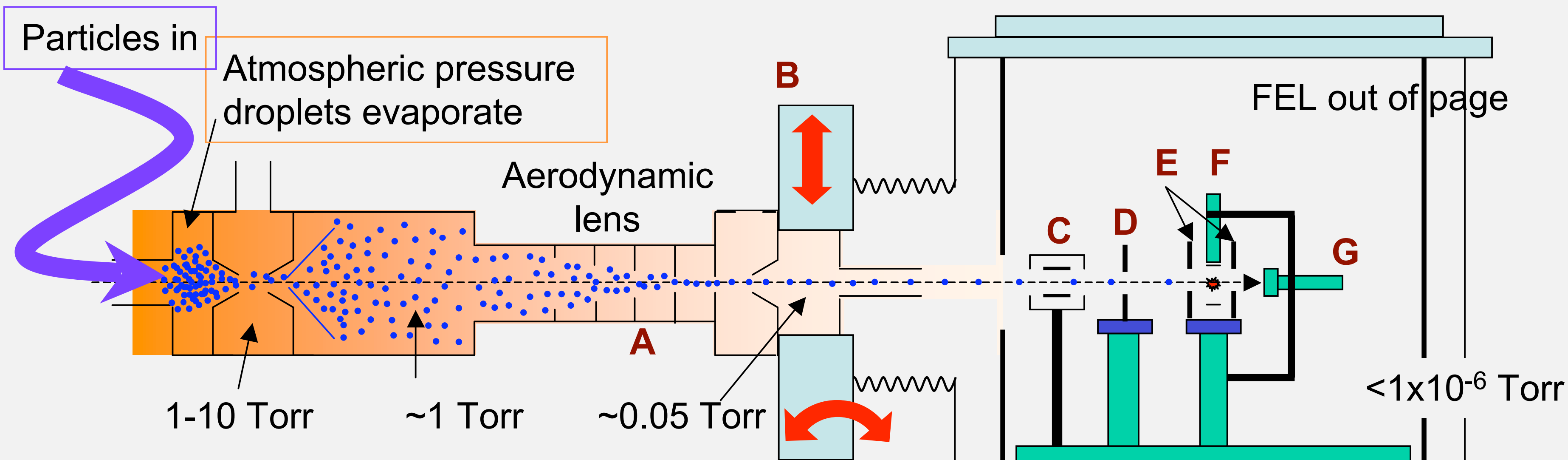
Picoplankton diffraction



Single shot diffraction of Cocolith



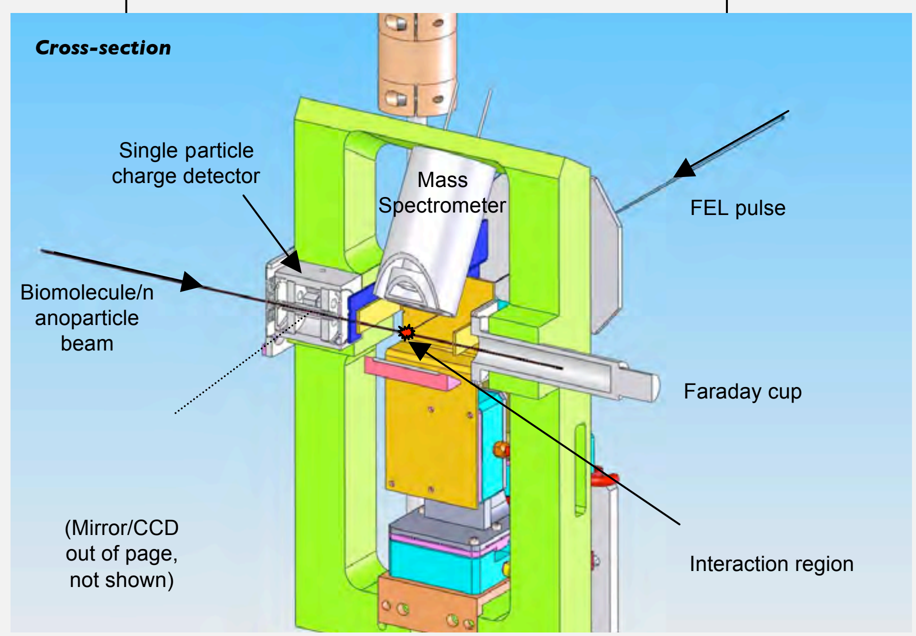
We have developed a particle injector for FLASH experiments



A. Aerodynamic lens
B. Particle beam steering
C. Charge detector
D. Particle beam skimming aperture
E. Particle beam alignment apertures
F. Time-of-flight mass spectrometer
G. Faraday cup

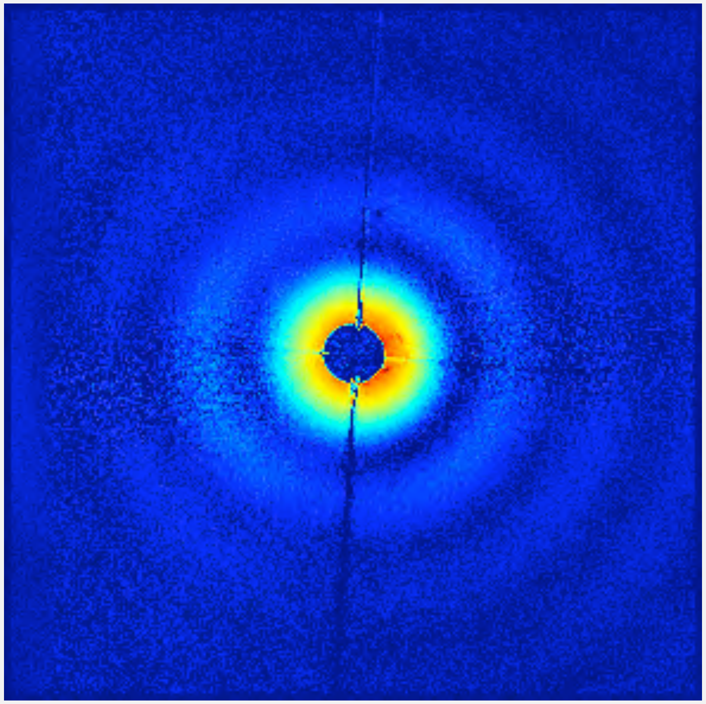
Measured particle flux:
88 nm spheres = 2.5x10^6 particles/second
28 nm spheres > 5x10^8 particles/second

On XFEL, predict about 1 Hz hit rate

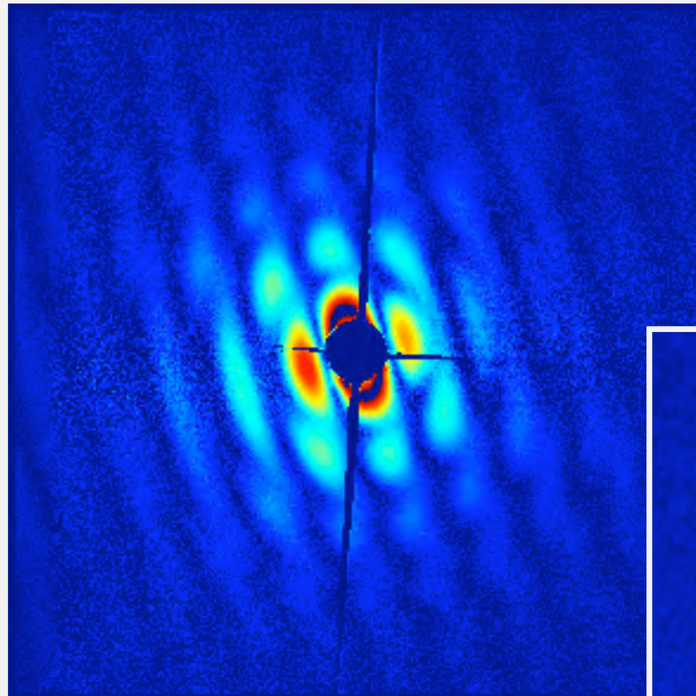


Single-particle FEL diffraction of “on-the-fly” particles has been demonstrated for the first time

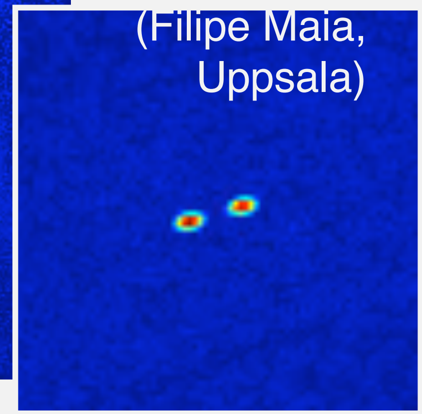
Single ~200 nm particle



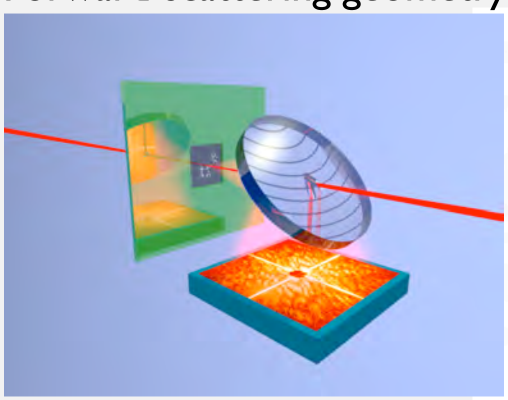
Two particles hit by the one pulse

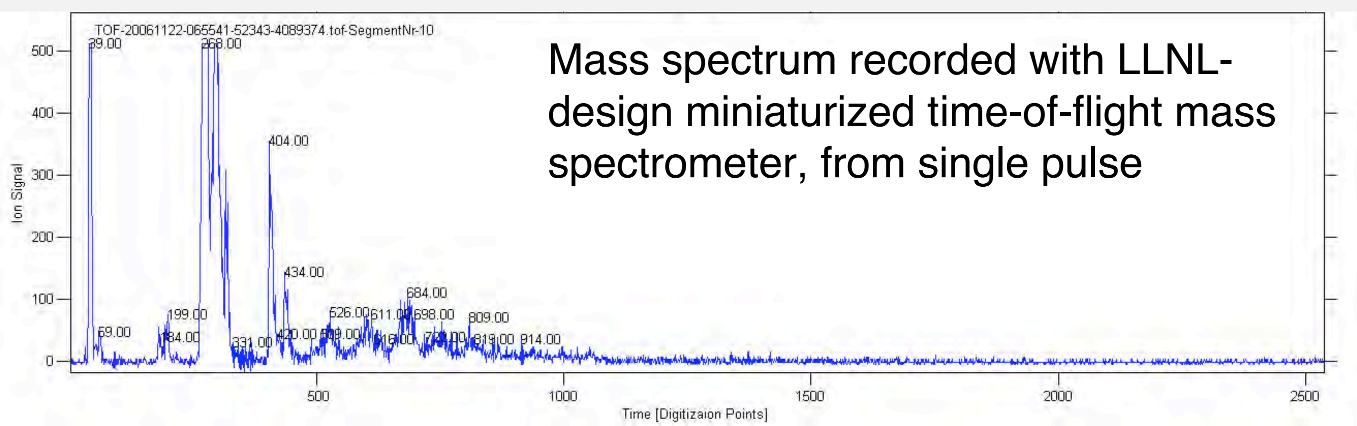


Reconstructed image (Filipe Maia, Uppsala)



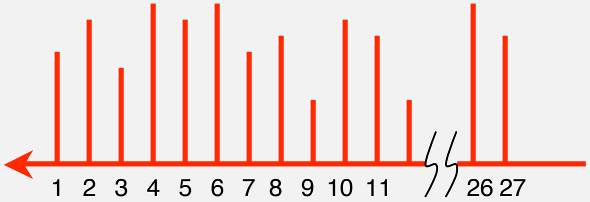
Forward scattering geometry



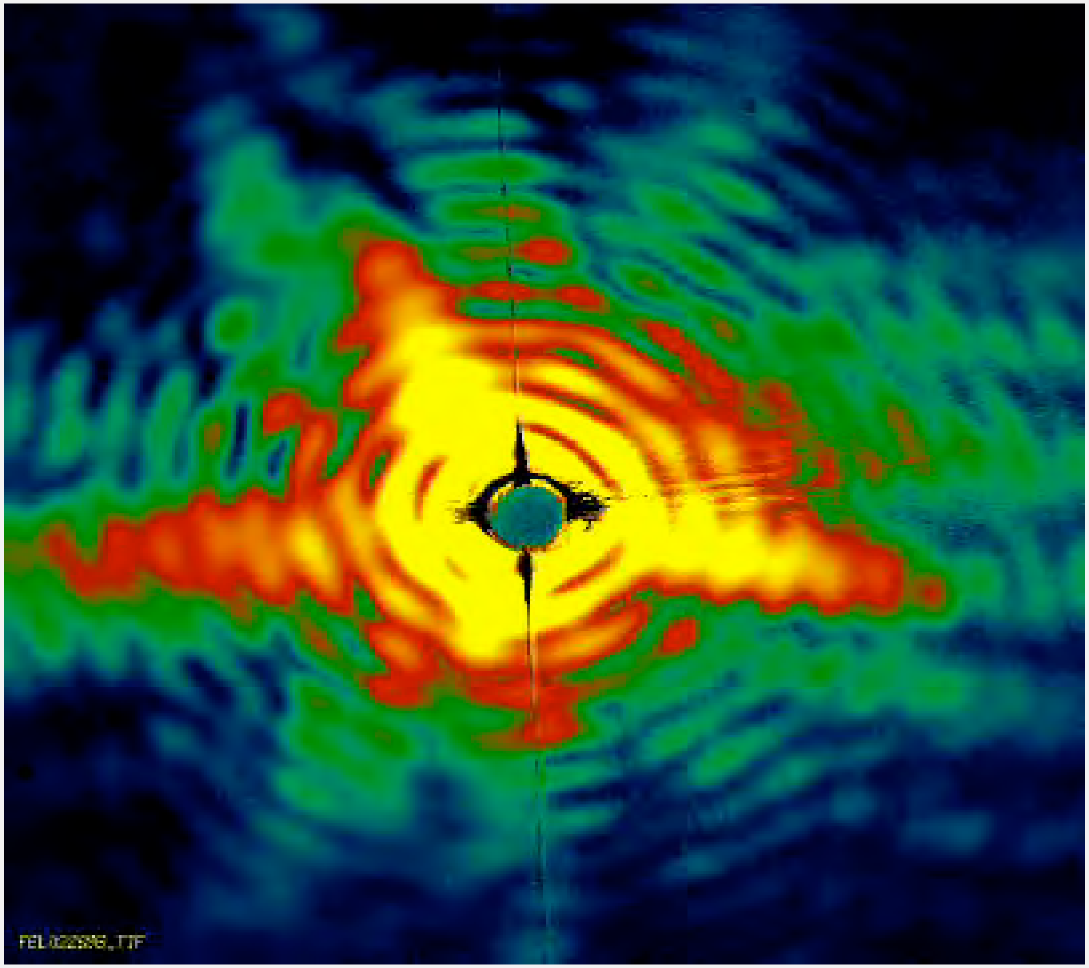


Mass spectrum recorded with LLNL-design miniaturized time-of-flight mass spectrometer, from single pulse

We achieved an acceptable hit rate for a wide variety of aerosol samples



10 fs FEL pulses at 10 μs spacing



2 sec exposures (played at 3 fps = 6x acceleration)
8 FEL pulse trains per exposure (5Hz FEL rep rate);
at 140 micobunches per pulse train = 1120 FEL pulses per image

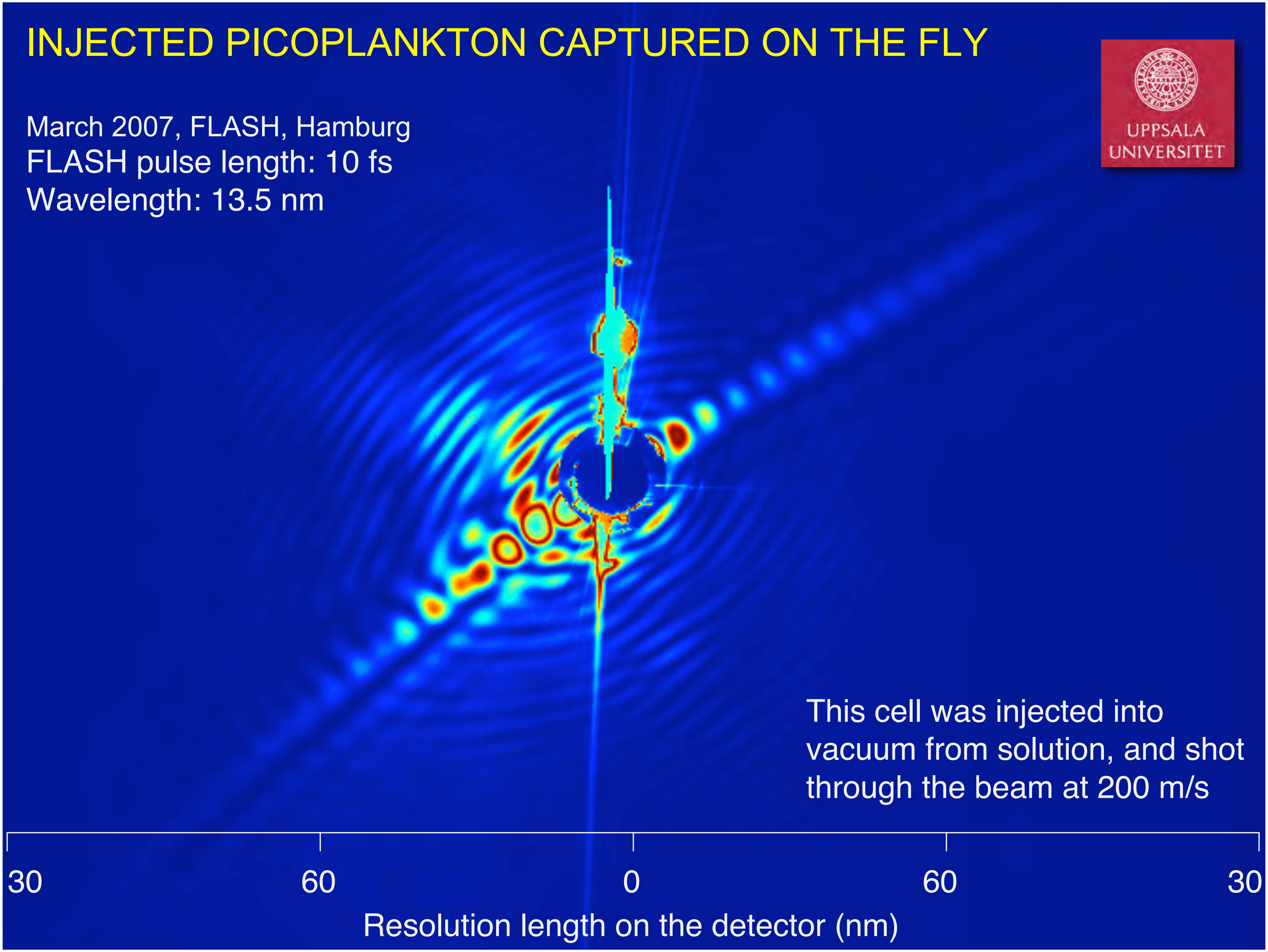
PICOPLANKTON **are the most abundant photosynthetic cells in the oceans (discovered in 1988)**

Phytoplankton account for nearly half of the total carbon dioxide fixed into the biosphere, while representing only about 1% of the total photosynthetic biomass on our planet. This is a most remarkable finding.

INJECTED PICOPLANKTON CAPTURED ON THE FLY

March 2007, FLASH, Hamburg
FLASH pulse length: 10 fs
Wavelength: 13.5 nm

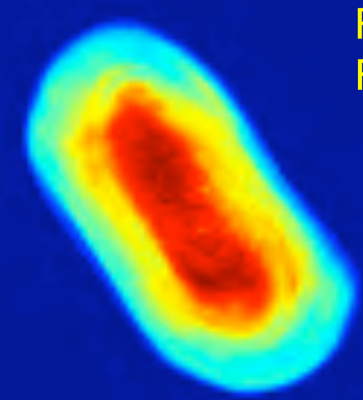
UPPSALA UNIVERSITET



This cell was injected into vacuum from solution, and shot through the beam at 200 m/s

FIRST RECONSTRUCTED IMAGE OF A LIVE PICOPLANKTON (CELL INJECTED INTO THE FEL PULSE)

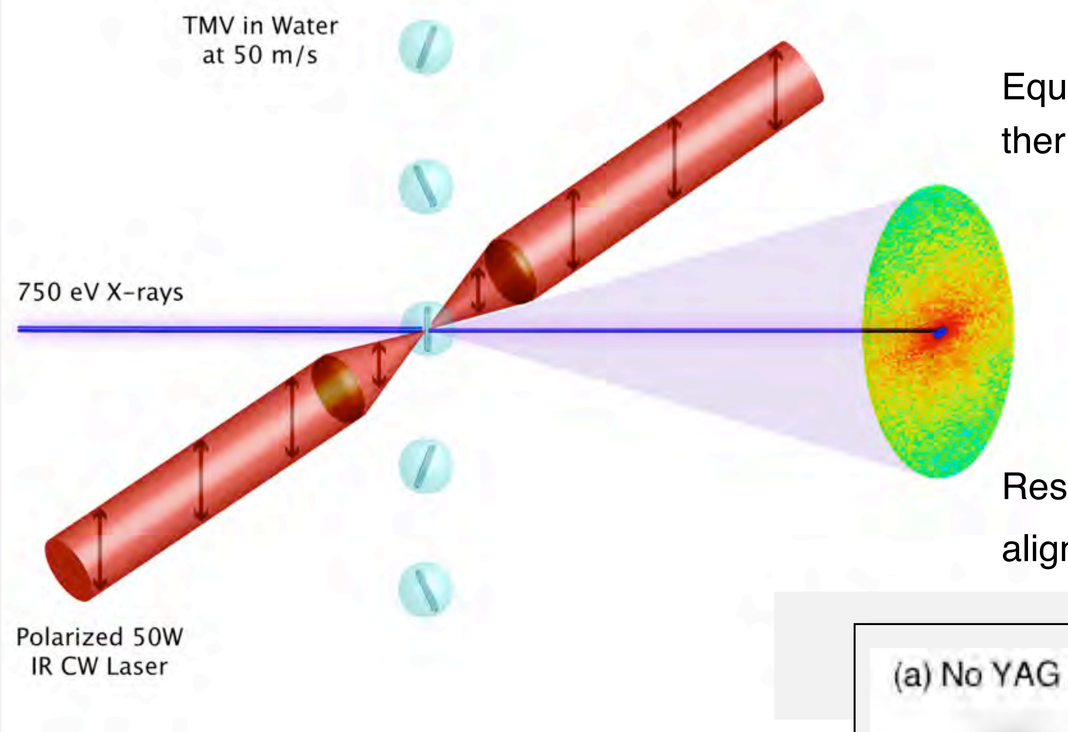
FEL pulse length: 10 fs
Wavelength: 13.5 nm



Reconstruction by
Filipe Maia, Uppsala

Reconstructed image of an intact picoplankton cell injected into the FEL beam

In the context of APS, laser alignment may make serial crystallography possible on a synchrotron (or ERL) source



Equipartition of rotational potential energy with thermal energy gives

$$\left\langle \Delta\theta^2 \right\rangle = \frac{T}{3\times 10^{-8}\, I \Delta\alpha}$$

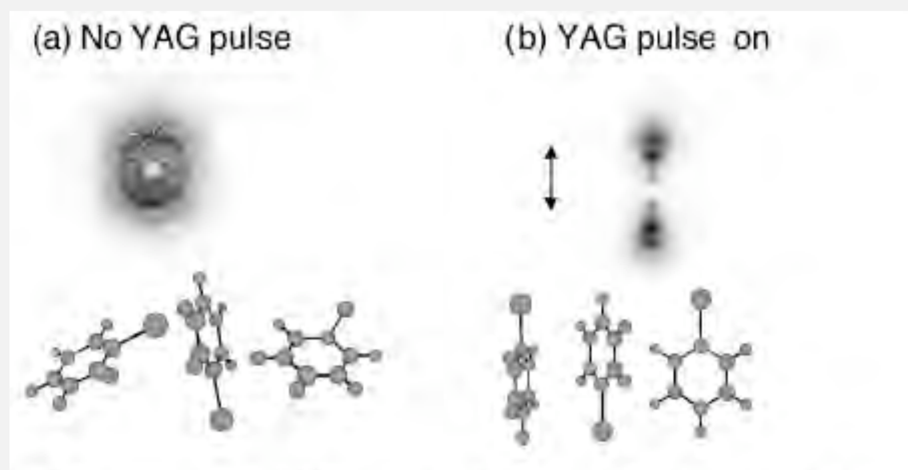
T - temperature in K
I - laser power in W/cm^2
$\Delta\alpha$ - polarizability anisotropy in nm^3

Resolution is limited by the degree of alignment: $d = (L/2)\,\Delta\theta$

*J.C.H. Spence and R.B. Doak, Phys. Rev. Lett. **92**, 198102 (2004)*

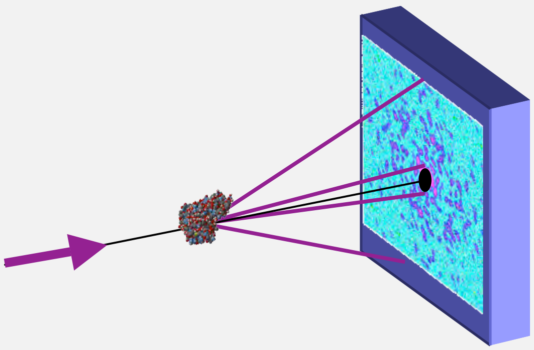
*J.C.H. Spence et al., Acta Cryst. A **61**, 237 (2005)*

*D. Starodub et al. J. Chem Phys **123**, 244304 (2005)*

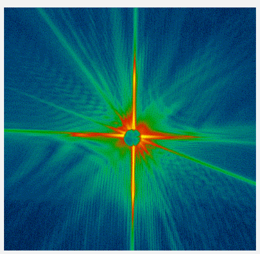


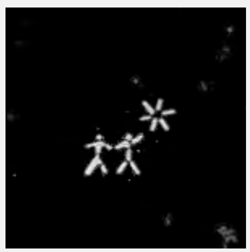
*Larsen, J. Chem Phys **111**, 7774 (1999).*

Overview

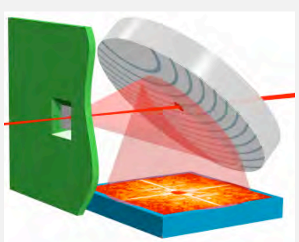


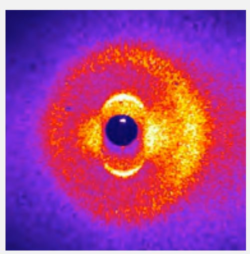
The ultimate goal of 3D diffraction imaging of single molecules is a ‘grand challenge’



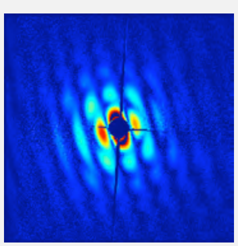


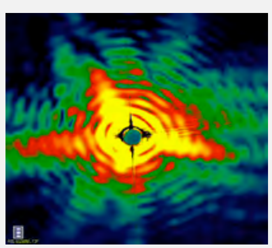
We have demonstrated diffraction imaging using single FEL pulses





Femtosecond X-ray pump-probe diffraction imaging can be used to image transient phenomena





We have demonstrated that we can image particles injected into the FEL beam ‘on the fly’